LOW TEMPERATURE CARBONISATION

LOW TEMPERATURE CARBONISATION

BY

C. H. LANDER,

D.Sc., M.I.Mech.E., A.M.Inst.C.E., F.Inst.P.

DIRECTOR OF FUEL RESEARCH UNDER THE DEPARTMENT OF SCIENTIFIC AND
INDUSTRIAL RESEARCH)

AND

R. F. McKAY

M.Sc., M.I.Mech.E., A.M.Inst.C.E.

PREFACE

THE subject of the low temperature carbonisation of coal is one which is attracting considerable attention at the present time; although it cannot be claimed that the difficult problems, involving both scientific and economic considerations, have yet been completely solved, there can be no doubt that considerable progress has been made in the direction of their solution.

The genesis of the book has been explained by Sir George Beilby in the Foreword. Its preparation proved to be of considerable difficulty as the subject is rapidly developing and many of the problems involved still require much further exploration. It was felt, however, that the subject could best be dealt with by giving the most reliable data on the different aspects of the work together with such conclusions as can safely be drawn from them. Unfortunately it is not always possible to draw definite conclusions and the Authors are only too well aware that, of necessity, some of the sections have been left with an incomplete survey of the aspects to which they relate. An attempt has been made to dissect the variables and to make each chapter self-contained so far as is possible. Such a method of treatment, however, inevitably involves a certain amount of repetition in a subject where such variables are so closely interrelated as in the present one.

The Authors have made use of material contained in official publications and have to thank the Controller of H.M. Stationery Office for permission to reproduce certain diagrams from these reports. Free use has been made of data contained in the proceedings of technical societies as well as of articles in the technical Press; where possible these have been acknowledged in the text.

The Authors wish to express their grateful appreciation of the advice and help which they have received from Sir George Beilby and for the interest which he has taken in the work; it is indeed to him they owe any special knowledge of the subject that may have justified them in undertaking the task.

They also wish to express their thanks to Dr. R. V. Wheeler and Dr. C. R. Young; to the former for much helpful criticism of the portions bearing on the constitution of coal and other aspects of the subject which he has made peculiarly his own, and to the latter for help and advice in the preparation of the sections bearing on the chemistry of the subject.

They are greatly indebted to Professor H. C. H. Carpenter, F.R.S., who has given advice on the properties of cast iron and other metals when exposed to high temperatures.

They are also indebted to Dr. Margaret Fishenden, Mr. J. G. King and other colleagues for much valuable assistance and to many firms and individuals who have supplied drawings and details of their processes.

In conclusion the Authors must pay a tribute to the generous manner in which Miss G. I. M. Blair has devoted her scanty leisure to preparing the manuscript for publication and for seeing the work through the press.

<div style="text-align: right">

C. H. L.

R. F. McK.

</div>

FOREWORD

It was suggested to me early in 1921 that a book on the subject of low temperature carbonisation would be welcomed by many who are interested in schemes for the more efficient utilisation of our coal resources. It is now widely recognised that this subject is of national importance, for if it becomes commercially feasible to carbonise in this way any considerable proportion of the tens of millions of tons of the coal annually consumed in the raw state in Great Britain, low temperature carbonisation would play a vital part in the solution of the smoke problem and in that of the home production of motor spirit and fuel oils. The subject, however, is far from simple, for its physical, chemical, mechanical and economic aspects are so closely interwoven that only after an intimate study of all these aspects ought any serious estimate of its development as a national industry to be attempted. In the absence of this all-round knowledge much that has been publicly spoken and written on this subject has been not merely futile but actually harmful to genuine progress.

In the Government Department of Fuel Research a carefully thought out scheme of inquiry has been steadily pursued for the past six or seven years. These inquiries embraced not merely the first-hand experimental work carried out at H.M. Fuel Research Station, but the similar work of outside inventors and experimenters all over the world.

The intimate association of Dr. Lander and Mr. McKay with the course and the results of these inquiries, and their complete dissociation from any interests, direct or indirect, in the commercial development of any particular schemes, placed them in an almost unique position as the Authors of a work on this subject.

With the full concurrence of the Department of Scientific and Industrial Research I therefore asked Dr. Lander and Mr. McKay to undertake this work. This they ultimately agreed to do, though not without considerable hesitation, in view no doubt of the heavy additional strain on their time and energy which would be involved.

Having now before me the work as a whole I feel that the pressure put on the Authors to undertake this exacting task has been fully justified. The more fully this work is studied the more evident will it become that the Authors have approached and dealt with their subject not only with great breadth of view but with a keen appreciation of the technical and economic problems which have been solved or are still awaiting solution.

G. BEILBY.

LONDON,
   March, 1924.

CONTENTS

CHAPTER I

INTRODUCTORY : AN INITIAL SURVEY

PAGE

CHAPTER II

THE AIMS OF LOW TEMPERATURE CARBONISATION

CHAPTER III

THE FUEL PROBLEM

CHAPTER IV

NOTES ON THE CONSTITUTION OF COAL

CHAPTER V

THE CLASSIFICATION AND TESTING OF COAL

CHAPTER VI

NOTES ON THE CHEMISTRY OF CARBONISATION PRODUCTS

CONTENTS

CONTENTS

CHAPTER X
OTHER TECHNICAL PROBLEMS

CHAPTER XI
THE PRODUCTS OF LOW TEMPERATURE CARBONISATION : COKE

CHAPTER XII
THE PRODUCTS OF LOW TEMPERATURE CARBONISATION : GAS

CHAPTER XIII
THE PRODUCTS OF LOW TEMPERATURE CARBONISATION : TAR

CHAPTER XIV
THE PRODUCTS OF LOW TEMPERATURE CARBONISATION : MOTOR SPIRIT

CHAPTER XV
NITROGENOUS PRODUCTS FROM COAL

CONTENTS

CHAPTER XVI

PROCESSES—EARLY WORK

CHAPTER XVII

PROCESSES—SOME LATER DEVELOPMENTS

CHAPTER XVIII

A DISCUSSION OF SOME DIFFICULTIES ENCOUNTERED IN LOW TEMPERATURE CARBONISATION

A. RETORT MATERIALS

CHAPTER XIX

THE ECONOMICS OF LOW TEMPERATURE CARBONISATION

LIST OF ILLUSTRATIONS

LIST OF TABLES

15

LOW TEMPERATURE CARBONISATION

CHAPTER I

INTRODUCTORY : AN INITIAL SURVEY

PHASES IN THE HISTORY OF LOW -TEMPERATURE CARBONISATION

THE history of low temperature carbonisation, so far as Great Britain is concerned, may be divided into two periods, the first dating from Parker's original proposals in 1906, and the second ten years later, beginning during the Great War, when the whole question of fuel utilisation proved to be of such importance that the subject of low temperature carbonisation was raised from relatively limited significance to a great national problem.

During the first period experimental work was pursued by Parker, Beilby, Maclaurin, and others. These investigators concentrated their attention chiefly upon the production of a smokeless fuel, and, during the earlier part of the period, regarded the crude oil obtained from their experiments as being of comparatively small importance. In 1912–13 the Royal Commission on Fuel and Engines for the Navy, under the chairmanship of Lord Fisher, exhaustively examined the possibilities of obtaining fuel oil from existing home industries. They reported that the only means of so doing lay in the "development of a new carbonising industry founded on the distillation of bituminous coal at a temperature much below that used in gas retorts or coke ovens." It was also clearly brought out in this Report that, as the possible oil production amounted only to some 5 to 8 per cent by weight of the coal treated, the creation of a market for the other products of carbonisation was necessary for the solution of the economic problem. The Report of the Commission naturally focussed the attention of the public on the whole subject of low temperature carbonisation, and not only provided a new incentive to investigation, but made the problem of the economical disposal of the coke assume new and huge proportions.

THE WORK OF THE FUEL RESEARCH BOARD

After the outbreak of the War the work of investigators remained in abeyance for a time, until the urgency of the supply of fuel oil in particular, and of fuel conservation in general, brought the definite recognition by the Government in 1916 that some measure of State Aid should be afforded in the solution of the many problems which presented themselves in this connexion. It was therefore decided by the Committee of the Privy Council for Scientific and Industrial Research in 1917, on the recommendation of their Advisory Council, to establish a Board of

Fuel Research, which should be responsible for the promotion and co-ordination of research into fuel. Sir George Beilby, who had been responsible for much of the experimental work and inquiries leading up to the Fisher Report, was appointed Chairman of the Board and Director of Fuel Research. The terms of reference of the Board were " to investigate the nature, preparation and utilisation of fuel of all kinds, both in the laboratory and, when necessary, on an industrial scale." After careful consideration of the whole fuel problem from the broadest point of view, the Board adopted as their immediate programme two lines of research, viz. :—

(1) A survey and classification of the coal seams in the various mining districts by means of chemical and physical tests in the laboratory.

(2) An investigation of the practical problems which must be solved if any large proportion of the raw coal at present burned in its natural state is to be replaced by the various forms of fuel obtainable from coal by carbonisation and gasification processes.

It will be seen, therefore, that under the second heading the problem of low temperature carbonisation naturally assumed a prominent place. At the time the Fuel Research Board was formed the shortage of fuel oil for naval and other purposes was reaching a maximum ; and it was pointed out to the War Cabinet by the Board that, although their schemes of investigation could have no possible effect in relieving the situation during the War, it was of paramount importance that this question of the supply of home-produced fuel oils for the Navy by the carbonisation of coal at present used in the raw state should be independently and exhaustively studied, " in order that a definite decision as to the possibility or impossibility of the proposals should once for all be arrived at."

The results of the work of the Board at H.M. Fuel Research Station at East Greenwich up to 1922 are given in their Report for the years 1920–21, published in 1922,[1] and a careful perusal of this document discloses the fact that, owing to economic changes which have come about as the result of the War, the solution of the problem has proved more difficult on the economic side than was anticipated. Much progress has, however, been made, and accurate data on many doubtful points have become available to all serious investigators. The Board affirm that much knowledge has been obtained in the direction of a technical solution, that this solution is almost within reach, and that the elements of uncertainty still remaining are mainly economic and social.

[1] Fuel Research Board. Report for the Years 1920, 1921 ; Second Section: " Low Temperature Carbonisation." (H.M. Stationery Office.)

STATE OF THE PROBLEM IN 1916

The rapid development of low temperature carbonisation has undoubtedly been greatly hampered by the tendency of some of the earlier pioneers to attempt the climbing of a long staircase at a single bound. Hence, when in 1916 attention was focussed on the problem through the shortage of fuel oil caused by the activities of German submarines, the whole subject was obscured by the many conflicting opinions and results obtained by the various groups of workers, whose most praise-worthy enthusiasm in some cases greatly exceeded their discretion. The Fuel Research Board, however, were able to select certain data of unquestioned accuracy as a basis for the prosecution of their own experimental inquiries.[1] These items were :—

(1) " The experience in Scotland for the past fifty years of the shale oil industry. This industry is founded on low temperature carbonisation, and about three million tons of shale per annum are distilled at low temperatures ranging from 450° to 550° C. for the production of paraffin oils and wax. "

(2) " The parallel experience of this industry in the carbonisation of bituminous coal for the production of fuel gas and other products.

(3) " The published records of the various steps in the development of Mr. T. Parker's proposal for the production of a smokeless solid fuel for domestic purposes by the carbonisation of bituminous coal in iron vertical retorts, of small cross-section, at a temperature of about 450° to 500° C. Mr. Parker's experiences established the soundness of his original contention that coke produced from bituminous coal at a temperature not exceeding 500° C., to which he gave the name of ' Coalite,' is an excellent and per-fectly smokeless fuel for domestic purposes. As he had prepared it, however, it was rather too fragile to stand the rough handling involved in transport to the consumer. Unfortunately, though large sums of money were expended on the testing and development of this method on a fairly extensive scale, success was not attained, and the problem of the establish-ment of a new carbonisation industry was still unsolved. The promoters of Mr. Parker's scheme appear to have then abandoned the use of metal retorts, and adopted firebrick ovens or retorts of the coke oven or gas retort type.

(4) " First-hand knowledge of the experiments carried out in the Maryhill works of the Cassel Cyanide Company, Glasgow, on the carbonisation of bitu-minous coal at temperatures ranging from 450° to 500° C. and on the briquetting of the coke so produced.

[1] *Ibid.*, p. 18.

(5) " Some knowledge of the experimental work of Mr. R. S. Richards on the carbonisation of bituminous coal at temperatures in the neighbourhood of 500° C. in an automatic machine designed by him.

(6) " Reports on the working of the Tozer retort for the carbonisation of bituminous coal and the production of smokeless solid fuel for domestic purposes.

(7) " First-hand information as to the experimental work of Mr. Robert Maclaurin in Glasgow on the carbonisation of bituminous coal by the sensible heat of a large volume of highly heated gas passed over and through a mass of broken coal in a vertical column or shaft of the blast furnace type."

At a somewhat later date official reports were available on two systems of carbonisation for the production of fuel oil which had been experimented with by the aid of Government subsidies. These were :—

(1) A retort of the Chiswick type which had been erected at the Nottingham Gasworks for the carbonisation of cannel coal.

(2) The " Barnsley " firebrick retorts, which had been subsidised by the Department of Explosives Supply with a view to the production of benzene and toluene. These retorts were subsequently abandoned and replaced by the setting described on page 206.

FURTHER ADVANCES SINCE 1916

From the foregoing, useful generalisations may be drawn, many of which have now been definitely and finally confirmed, as the result of the work of the Fuel Research Board. Of these, the selection of suitable coals appears to be by far the most important. Parker's original iron retort possessed many excellent features, but broke down through the use of unsuitable coal, and an excessive heat gradient from the furnace or oven inwards. The importance of the rich gas as one of the most desirable assets of low temperature carbonisation has only been recognised comparatively lately ; and, whatever were the other defects possessed by the Parker metal retorts, they were at any rate perfectly gastight, a feature which long experience in the Scottish shale oil industry has shown to be unshared by any type of firebrick retorts worked under temperature conditions corresponding to those necessary for the production of smokeless fuel and oil.

' The preparation of the coal for carbonisation has also been shown by the Fuel Research Board to be of immense importance in the production of a robust coke. The material, which even in a single coal consists of bands of varying fusibility, must be broken down and intimately mixed, so that the properties of the more

fusible portions shall be utilised to bind together the less fusible particles ; and, if the coal is deficient in either of these two constituents, the correct proportion must be ensured in some way, usually by blending in suitable proportions with another coal possessing the properties in a high degree which are deficient in the first.

The question of drying the coal is also of some importance. The operation of drying is much simpler than that of carbonisation ; and it is obviously uneconomical both in capital and heat to use an expensive retort for an operation which can be performed much more cheaply in an apparatus especially designed for the purpose.

Another important reason for the failure of some of the earlier processes lay in the tendency of the inventor to attempt too much in the effort to produce an apparatus of universal application to shales, cannels and coals. Such investigators would have been well advised to select one material for experiment, and to continue work upon that material until success had been attained. On the contrary, however, retorts were erected which, long before they had proved themselves in any direction, were provided from day to day with coals of widely differing characteristics. Under such conditions no retort, however excellent, had a reasonable chance of being perfected, and the results obtained, although imposing, possess little value to the discerning mind.

As a result of the experimental work of the Board, it is to be hoped that the time has gone by when the costly mistakes which have been made in the past will be repeated. Although it cannot be claimed that complete success has been achieved, data of unquestionable accuracy are at least now available for all serious investigators working on the difficult problem of low temperature carbonisation.
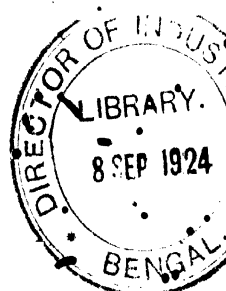
CHAPTER II

THE AIMS OF LOW TEMPERATURE CARBONISATION

THE IMPORTANCE OF COAL

THE importance of coal as one of the chief sources of our national wealth cannot be disputed ; nor, in the face of our dwindling supplies and the increasing difficulty of winning these supplies, can it be said that too much attention is being paid to the problem of the scientific utilisation of our coal reserves. In the past, fuel has been used by man in ever-increasing quantities ; but in most parts of the world the supply has been so plentiful compared with the requirements that there has been little inducement until recently to study the most economical methods of its use. The conception, at one time of popular acceptance, that coal is a fuel which will mainly be consumed in the raw state, is at length being replaced by the correct and scientific view that coal is a commodity which should be made to yield other fuels of higher " availability," and from which valuable by-products as well as fuel should be obtained. Although scientifically the burning of " raw " coal in boilers and furnaces for the production of heat and power must be deprecated, it must also be recognised that no adequate substitute is yet forthcoming. Water power on a large scale is unobtainable in this country ; and, while it is to be expected that an increasing and extended use will be made of oil fuels, this country is entirely dependent upon imported supplies. The present demand for oil fuel is already beginning to encroach seriously upon the estimated oil reserves of the world, and unless or until vast fresh oil fields are located, or new sources of supply found, there is no possibility of the supplanting of coal by oil as the main source of our power supply. As for gaseous fuel, the total substitution of gas for the raw coal at present consumed in producing heat and power introduces problems which for various reasons are not easy of solution. In any case, the extension in the use of both oil and gaseous fuels leaves untouched the pressing problem of the supply of a satisfactory fuel which can be burnt in the already existing domestic grates without involving much, if any, cost on account of their structural alteration.

THE CARBONISATION OF COAL

It is well known that the carbonisation of coal, i.e. the heat treatment without access of air, yields solid, liquid and gaseous products—solid fuel in the form of coke, liquid fuel such as motor spirit, fuel oil, lubricants, etc., gaseous fuel suitable for lighting, power and heating purposes, together with other products which may be used in the preparation of fertilisers, explosives, disinfectants, or which form the basis of the artificial dye industry. This treatment of coal is not only desirable in order to increase the " availability " of the potential thermal units in the coal

22

—a term which will be further explained in the next chapter—but also to recover valuable by-products such as sulphate of ammonia, etc., which are not fuels.

The carbonisation of coal is much more complex than is the distillation of a simple liquid mixture. In the latter case distillation may be carried out in stages, and the resulting fractions can in general be readily separated. In other words, the distillation process is purely a physical phenomenon. In the case of coal, however, chemical reactions may proceed simultaneously with physical modifications as the temperature of the material is increased, and different products may be evolved, the separation of which is a tedious and costly proceeding. Further, the final form of existence of the products yielded by the carbonisation of coal is influenced by considerations other than the temperature employed. Not only is the raw material itself a variable factor, but the conditions under which the process is carried out also affect considerably the final products of distillation.

There are no definite lines of demarcation between the various methods whereby coal can be carbonised, and the temperatures employed merge gradually in different types of plants from high, medium to low. Again, it may be found that a process of carbonisation, first at low temperatures and afterwards at high temperatures, may in certain cases give results of economic value. The process of liquefying certain coals by hydrogenation under pressure, into which considerable research is being made at the present time, also points out a method of treating coal which may possibly in the future prove a rival to those processes at present practised.

No attempt will be made here to adjudicate upon the controversies between the advocates of the different methods of carbonising coal, each of whom would naturally like to see the process in which he is interested become of universal application. There is, however, ample room in industry for a variety of processes, since the distillation of coal yields a large number of products, and it must be recognised that the maximum quantity and the highest quality of each cannot be obtained simultaneously. Thus the main desideratum of high temperature distillation in gas retorts or by chamber carbonisation is a maximum yield of gas of the " declared " calorific value, and the recovery of coke, tar, etc., must be termed of secondary importance, although the financial results from the sale of these by-products play an important part in making the process successful commercially. In the by-product ovens of the Koppers or Semet-Solvay type, gas and tar are in effect by-products, and a maximum yield of hard metallurgical coke is of primary importance. In producer gas plants a supply of gas for power purposes—not necessarily of high calorific value—is desired. The main desiderata of low temperature carbonisation are, however :—

(1) The production of smokeless fuel suitable for domestic and other purposes.
(2) The maximum production of oil of low specific gravity.
(3) The maximum production of gas of high calorific value.

The amount of incondensable gas which is obtained in low temperature carbonisation is comparatively small, but as the gas possesses special properties and has a high calorific value, its sale will have an important bearing on the financial success of the process.

The volatile content of the coke varies from 8 per cent to 10 per cent against a volatile content of 0·1 per cent to 4 per cent for high temperature coke. Low temperature coke can be easily ignited, and can be used in open fire-places as at present constructed.

SMOKE POLLUTION

The wasteful practice of burning raw coal in furnaces and domestic hearths not only represents a direct national loss through the non-recovery of valuable by-products from the coal, but is the chief cause of the smoke nuisance of large cities and manufacturing centres. The pall of smoke which hangs over all industrial

TABLE I

DEATH RATE AND CAUSES OF DEATH IN LONDON, IN NOVEMBER, 1922

Week Ending	DEATH RATE.		CAUSE OF DEATH IN LONDON.		
	London.	England and Wales.	Bronchitis & Pneumonia.	Heart Diseases.	Other Diseases.
Nov. 11, 1922 . .	12·5	12·6	208	198	677
,, 18 . .	13·3	12·9	257	219	679
,, 25 . .	14·3	12·6	326	265	689
Dec. 2 . .	13·0	12·6	208	181	680

towns on a calm day is a sufficient indication of the attendant evils. At these great centres, where large numbers of people must necessarily reside, public health and plant life not only suffer from the ensuing pollution of the atmosphere, but also from the reduction of the hours of available sunshine. Thus the figures in Table I are significant when it is remembered that, early in November, 1922, London was visited by a severe fog. It will be seen from the table that, whereas the death-rate over the country remained approximately constant throughout the month, that in London was considerably increased, due chiefly to deaths from bronchitis and pneumonia. Other factors must of course be taken into consideration ; but that smoke pollution is a contributory cause to general ill-health appears to be clearly indicated not only from this table, but from other statistics bearing on this particular subject.

Fogs not only affect adversely the health of the people, but cause vexatious

delays in traffic and loss of time and temper, while involving great expenditure on artificial illumination. Furthermore, public buildings and monuments are discoloured and damaged by the soot and acid of the smoke. Even in the average household the additional expense of the smoke nuisance is appreciable. An investigation was made in 1918 to ascertain the comparative cost of household washing in Manchester and Harrogate. It was found that the extra cost in Manchester for fuel and washing material, attributable to the smoky atmosphere, was equivalent to about 7½d. per week per household. It is computed that the total yearly loss for Manchester and Salford is nearly half a million sterling. The baneful effects of smoke in cities are fully described in the Final Report of the Committee on Smoke and Noxious Vapours.[2]

Smoke is emitted to the atmosphere in the main from two offenders—the industrial and the domestic chimney. Their relative culpability differs in various localities, and the remedy in each case is not the same. The employment of mechanical stoking and careful hand stoking has, in the case of industrial plants, done much to render possible that constancy of fuel supply to boiler furnaces which prevents the formation of smoke ; while the higher temperature of a well-regulated furnace over that of a domestic fire tends to make any resulting smoke less obnoxious and injurious. It is generally agreed that, at least in many cities, the domestic chimney is the chief offender as regards the smoke nuisance.

Little can be done locally to reduce the smoke nuisance from the domestic chimney. Sir Napier Shaw,[3] the Chairman of the Advisory Committee on Atmospheric Pollution, has suggested that a reduction in rateable value should be made for a smokeless house. Moreover, in a large number of the houses now being built in various parts of the country provision is made for only one coal fire per house— that is, one in the main living-room. It should be further noted that in the recently published report of the Public Control Committee of the London County Council the Committee express the opinion that the time has arrived when the use of the old-fashioned kitchener might well cease ; not only because it is inefficient, but also because it must be held responsible for a very large proportion of smoke arising from domestic chimneys.[4]

Suggestions of this description are, however, not always immediately practical, and in any case are only palliatives. The smoke nuisance problem must be examined from the broad national standpoint, and its solution will be hastened if it is found possible to obtain an adequate supply of smokeless fuel suitable for domestic purposes. Undoubtedly the burning of smoky coal for domestic purposes cannot

[1] Thomson, W. *Manchester Guardian*, 22nd February, 1922.

[2] Ministry of Health. Committee on Smoke and Noxious Vapours Abatement : Final Report. (H.M. Stationery Office.)

[3] Shaw, Sir Napier, F.R.S. *Times*, 23rd August 1921.

[4] London County Council : Report of Public Control Committee, 3rd July, 1923.

well be prohibited by legislation until a recognised valuable and economic substitute for raw coal is upon the market in sufficient quantities.

SMOKELESS FUEL

The use of gas for cooking purposes is diverting a certain proportion of coal from the domestic fire-place, but neither gas nor electricity seems likely to become universal for heating purposes in households. Although the efficiency of a domestic fire-place as a heating agency for rooms is low, it must be recognised that factors other than thermal efficiency render it improbable that open fire-places will be superseded at an early date. The average Englishman likes to see a cheerful fire in his living-rooms, and this cherished desire is proof against such scientific details as lack of efficiency, etc. Although national sentiment in favour of open fire-places is not likely to be changed in the near future, many of the drawbacks of the present system would be reduced by the substitution of a good smokeless fuel for the soft bituminous coals now used. There is therefore ample scope for the development of suitable processes for the carbonisation of coal at low temperatures, and it is reasonable to expect that commercial success will eventually be attained.

The requirements of a good smokeless fuel in this country may be thus briefly summarised:—

(1) It must have been previously treated for the recovery of valuable by-products from the raw fuel and so rendered smokeless.

(2) It must contain sufficient volatile matter—say 7 per cent to 10 per cent—or be of such a structure as to be easily kindled and kept alight in open fire-places as at present constructed, i.e. it should require very little draught for combustion.

(3) It must have a relatively low ash content ; partly to prevent an undue reduction in its calorific value, and partly to reduce the dust resultant upon combustion when burned in household fires, and to reduce clinkering troubles when burned in boiler furnaces.

(4) It must not be so friable as to break easily during handling and transport.

(5) It must be compact, but not of such a structure that the ash formed during combustion covers the surface of the fuel in such a manner as to hinder combustion or to mask radiation.

(6) Its price must be sufficiently low so that when the other advantages are taken into account it will attract purchasers away from bituminous coals. If the coke be sold in the form of briquettes, the advantages of uniformity and coherence should be taken into account when considering the economic value of the briquettes.

OIL FUELS

According to Lloyd's *Register*, the s.s. *Bakuin* was fitted for burning oil fuel in 1892, and was in all probability the first oil-burning steamer engaged in continuous sea service. Since that time engineers have watched with interest the great developments which have taken place in the substitution of oil for coal in ocean-going vessels. During the past fifteen years the gradual replacement of coal by oil has been going on in the Navy, and at the present time almost complete substitution has been effected.

In the Mercantile Marine developments have been less rapid, owing to the fact that in this service overall economy in working must necessarily be a consideration of greater urgency than in the Navy, where the efficiency of a ship as a fighting unit is of the first importance. It is, however, worthy of note that in the " Report of *Lloyd's Register of Shipping* for the Year 1922-3 " it is stated that " vessels to the number of 123, representing 782,830 gross tons, or 48·4 per cent of the total tonnage of new vessels classed during the year under review, were fitted for burning oil fuel." At the same time it should be mentioned that the Society's Report points out that it does not necessarily follow that all such vessels are at present using oil. Again, since July, 1914, the tonnage of vessels fitted for burning oil fuel on *Lloyd's Register* had increased almost twelvefold by July, 1923, and vessels fitted with oil engines had increased from 297 (234,287 gross tons) to 1,831 (1,668,414 gross tons) in the same time.

Even before the great European War the question of the home supplies of oil fuel for the Navy and Mercantile Marine had been given anxious consideration by those in authority. The subject was first exhaustively examined by Lord Fisher's Commission during 1912-13, while emergency inquiries were conducted during the War with the same object. None of these later inquiries added materially to the information which had been collected and verified by the Fisher Commission, whose main arguments are as true to-day as they were in 1912 ; while, as we have just shown, the urgency of the problem has been greatly intensified.

The importance of this matter was demonstrated most forcibly during the War, for it is generally known that the reserves of oil in this country were on occasion dangerously low. The only practicable steps to relieve this shortage from home sources that could then be taken were to develop the Scottish shale industry to the utmost, and to divert for Admiralty use as much as possible of the heavy oil produced by the existing carbonising plants. The augmented supply of fuel oil from these sources was, however, very small relative to the war demands of the Fighting Services. Although the requirements of the Navy have now been reduced to pre-War standard, or even below it, the increase in the use of oil in merchant vessels makes the question one of rapidly increasing importance from the national

point of view. It is true that the period of scarcity and high prices has passed for the present, but the fact still remains that this country is dependent upon overseas supplies for all fuel oil required. The amount of fuel oil that can be obtained by the carbonisation of coal at the high temperatures employed in ordinary gasworks or coke ovens is comparatively small, even apart from the fact that there is already a regular outlet for the products ordinarily obtained by these processes. There remains, however, the possibility of employing some process of carbonisation at low temperatures, and the position thereby created is summarised in the Report of the Fuel Research Board for the years 1918–19,[1] as follows :—

> "If bituminous coal is to become an important source of home supplies of fuel oil for the Navy and Mercantile Marine, tens of millions of tons per annum of coal, which is at present burned in the raw state, would need to be subjected to a preliminary process of carbonisation, and thereby split up into gaseous, liquid and solid fuels, which collectively would be used to replace this raw coal for the purposes for which it is at present consumed."

It must be recognised that the value of the oils obtained by low temperature carbonisation will be dependent for many years to come on the competitive price of the petroleum obtained from the oilfields of the world. This value will rise and fall for reasons outside the control of those in charge of low temperature processes, and may possibly fluctuate independently of the markets for the other commodities. Until the industry has become stabilised, therefore, optimistic monetary values of low temperature oil should be accepted with caution.

LOW TEMPERATURE GAS

In the early attempts to work low temperature processes the gas obtained was used to supply the heat required for the retorts. It must be said in passing that, owing in some cases to loss of gas through the use of unsuitable retort materials, the heat so obtained was insufficient for the purpose, and extra sources of heat had to be supplied. The passing of the Gas Regulation Act of 1920, whereby gas companies are empowered to charge for gas by calorific value instead of by volume, has, however, placed a new value upon the gas obtainable by low temperature carbonisation. This gas, being very high in calorific value if uncontaminated by the heating medium, would be exceedingly useful as an enriching agent for increasing the calorific value of low grade gas, and is therefore of direct interest to the gas companies. It is almost certain that improved economic results will be obtained by the use of producer gas of low calorific value as the heating agent for the low temperature retorts and the sale of the rich gas obtained by the

[1] Fuel Research Board. Report for the Years 1918, 1919. (H.M. Stationery Office.)

process. This, as was first pointed out by Sir George Beilby,[1] may form a connecting link between the gas-making industry and the establishment of the new process, and the gas may become one of the most valuable and least speculative of the assets of low temperature carbonisation.

RIVALRY BETWEEN CARBONISING SYSTEMS

It is perhaps not difficult to appreciate the want of enthusiasm which has in the past been shown by gas engineers towards the process of low temperature carbonisation. Not only are the processes of high and low temperature carbonisation very distinct, but in the former case we have an industry which is, after years of experience and development, well established, in which many millions of pounds have been invested, in which trustworthy data relating to processes are published in numerous technical papers, and in which the values of the products of carbonisation are well known and where these products are widely utilised in industry and in the home. In the latter case we have an industry, or a possible industry, which has not yet been placed upon a stable financial basis, in which there has not until lately been available any independently established data, in which much research work has yet to be undertaken, and in which markets must be created for the products of carbonisation.

The rivalry between the advocates of each system of coal carbonisation is commercial rather than scientific. Each system may supply a want which the other cannot satisfy, and one cannot therefore under present conditions displace the other. Uniformity of treatment of all our coal supplies by any one process could only be possible if all coal were alike and there were no demand for the products of other processes. The question of the relative efficiency of different processes, though interesting from a scientific standpoint, is therefore not of primary importance.

DIFFICULTIES IN THE ESTABLISHMENT OF LOW TEMPERATURE CARBONISATION

It is unnecessary to enlarge upon the great possibilities of the products of low temperature carbonisation. These have already been put forward in recent years, with perhaps more than justice, in the statements which have been made by sanguine inventors, and in the prospectuses of companies which have been floated to develop one or other patented process. It has been truly said that the advantages of low temperature carbonisation are too obvious. Many of the excellent results obtained from materials in the laboratory are unquestioned, but before the same results can

be produced by a plant on a commercial scale many technical problems have to be solved and much large-scale experimental work undertaken. The necessity for technical investigation and control cannot be overestimated, and it will be found in this, as in so many other manufacturing enterprises, that favourable results are largely influenced by the many small economies and modifications of working determined by careful supervision. The probability of the commercial success of any low temperature process and its capability of general application cannot, indeed, be determined until after full-scale plants have been in operation for considerable periods.

The process of coal carbonisation itself is most complicated, and the problem is rendered still more complex by the wide differences which exist between different coals and even sometimes between samples of coal taken from the same seam. Proximate and complete analyses, though useful, are not complete indications of the behaviour of coal in all types of retort, and microscopical research work has not yet been fully co-ordinated with the results of actual experience. In order to assess any low temperature process commercially, not only have the kind of material treated and the products of distillation to be considered, but also the methods of operating the plant and the mechanical details of the retort. Although much work has been done to establish the broad underlying physical and chemical principles governing low temperature carbonisation, a more detailed amplification of these principles to suit local conditions has still to be determined.

Broadly speaking, the establishment of low temperature carbonisation as a commercial possibility is dependent upon four important factors, viz.:—

(1) There must be ample supplies of the raw material.

(2) The plant must be such as will have a reasonable life, and it must be under the control of an experienced and scientific staff.

(3) The products of distillation must be in a marketable form, either for the direct use of consumers, or as the raw material in other industries.

(4) The commercial value of the products after retorting must admit of a reasonable margin of profit after meeting the working costs and manufacturing expenses, including the depreciation and renewal of the plant.

THE ESTABLISHMENT OF MARKETS FOR LOW TEMPERATURE PRODUCTS

While the four factors given above will be discussed in greater detail, there is one aspect of the third which might conveniently be enlarged upon at this stage, viz. that there does not at present exist a demand for the special products of low temperature carbonisation. A market there assuredly is for coke, oil and gas

obtained by low temperature carbonisation; but it is a market in which the special properties of these products receive little, if any, commercial recognition. For example, there has been sufficient low temperature coke distributed and sold within recent years for its admirable properties as a domestic fuel to be widely recognised. There is, however, as yet no great demand amongst householders for this fuel in preference to raw coal; and, if it were produced on a large scale, the degree to which its superiority over raw coal would be reflected in the price which consumers would be willing to pay is unknown.

Low temperature oil and low temperature gas are in the same category, although not to the same degree. It will be found later that the economic stabilisation of low temperature carbonisation is largely dependent upon the establishment of markets for the gas, coke and oil, in which there is a commercial recognition of the special properties of the products obtained by the process. Until a full-scale plant has been running for some considerable time and has been disposing of its products in a free market, any enhanced estimates of the value of those products must be accepted with caution.

CHAPTER III
THE FUEL PROBLEM

ASPECTS OF THE FUEL PROBLEM

THE fuel problem may be stated in many diverse ways. It may be that, in the first place, it is desired to draw attention to the world's diminishing supplies of the known fuels, to the increasing difficulties in obtaining those supplies and to the corresponding necessity for utilising them economically and efficiently; it may be that, in the second place, it is desired to urge the necessity of research for additional or alternative supplies of fuel; or it may be that, in the third place, it is desired to study the methods whereby householders and manufacturers may obtain heat and power from fuel in the most convenient form for their immediate needs.

Many papers have already been written on the first statement of the problem, and it is only necessary here to recapitulate some of the main facts. The second statement of the problem falls outside the scope of this volume, but it is desirable to discuss fully aspects of the third statement of the problem with particular reference to the process of low temperature carbonisation.

COAL

In regard to the first statement of the problem, it is recognised that coal is the most important source of the world's fuel supply. The amount of the world's coal reserves is, however, a very hypothetical figure. At the World's Geological Congress held in 1913, these reserves were estimated to be about 7.4 million million tons. Of the total, the reserves of the United States were estimated to be about 52 per cent, of Canada about 17 per cent, of China about $13\frac{1}{2}$ per cent, of Germany about $5\frac{3}{4}$ per cent, and of Great Britain only about $2\frac{1}{2}$ per cent. It is not necessary here to discuss the validity of these figures, or the probable length of time required to exhaust these stores, though it should be noticed in passing that the various estimates made by experts agree that this period of time must be measured in hundreds of years only. A more important matter from the point of view of this country is the comparison of the coal supplies of Great Britain with those of her nearest trade rivals. One of the reasons for the great increase in the prosperity of Great Britain during the past century was the plentiful supply of good and cheap coal. This enabled manufacturers to keep down the costs of production and gave them opportunities of competing successfully in the world's markets. It is now agreed, however, that, at the present rates of consumption, the reserves of Great Britain will be exhausted long before those of America and Germany. Although this is by no means an urgent matter as yet, it can be seen that, as our resources dwindle, the cost of winning the coal from greater depths and from thinner seams must increase, so that it may be only within a few generations that our coal exports

will diminish to a negligible quantity, and our industries will be penalised by paying more for their coal supplies than do the industries of their foreign rivals. It is the price of coal, and not the quantity of the reserves, which governs the commercial utility of our national coal resources. Though the reserves in Great Britain are not likely to be exhausted for many years, the present generation must be considered the guardians of the geological heritage it has received.

The world's production of coal at present is of the order of twelve hundred million metric tons. Table II gives the amounts produced in various countries for the years 1919, 1920 and 1921, and also the information received by the United States Geological Survey up to March, 1923, on the world's coal production in 1922. The production of certain countries for 1922 is not given, but the total of the missing data ordinarily amounts to 4 per cent only of the aggregate. The estimated total for 1922 given in Table II is not likely, therefore, to be very different from the actual production. The unit used is the metric ton of 2,205 lb., the approximate equivalent of the long or gross ton.

TABLE II

ESTIMATE OF THE WORLD'S PRODUCTION OF COAL IN THE CALENDAR YEARS 1919, 1920, 1921, AND 1922

Country.	1919.	1920.	1921.	1922.
Australia	10,736,321	13,176,426	13,073,845	*
Belgium	18,482,880	22,388,770	21,807,160	21,234,170
British India	22,991,217	17,356,889	• 19,511,154	19,000,000
Canada	12,411,328	15,088,175	13,555,462	12,890,468
China	18,292,252	19,484,896	*	*
Czecho-Slovakia	26,946,813	31,086,479	32,699,111	29,000,000
France	22,341,000	25,300,000	28,976,495	32,000,000
Germany—Coal	†116,707,200	†140,757,433	†145,610,000	†130,965,000
,, Lignite	93,645,500	111,631,000	123,011,000	137,207,000
Japan	32,703,338	30,819,898	26,000,000	25,000,000
Union of S. Africa	9,313,232	10,408,497	10,331,886	10,000,000
United Kingdom	233,467,478	233,216,071	166,922,000	255,892,000
United States	502,534,410	597,168,000	459,402,000	417,000,000
Other Countries	46,553,865	49,068,527	*	*
	1,168,000,000	1,317,000,000	1,133,600,000	1,200,000,000

* Estimate included in total.   † Includes Saar and Upper Silesia.

Of the major coal-producing nations, Great Britain and France are therefore the only ones to show an increase for 1922 over 1921. The proportion contributed

by the United States was about 35 per cent of the total, and the reduction in output during the years 1920-2 is very marked. The proportion contributed by the United Kingdom is just over 20 per cent of the total, a great increase over that of the preceding year ; but the drop in the production for 1921 must be attributed to some extent to the coal strike of that year.

The proportion contributed by Germany, including the output of lignite, is just over 22 per cent of the total. According to the Report of the Reich Coal Council, the German production of coal for 1922, exclusive of the Saar district, amounted to 129,964,000 tons, which is about $4\frac{1}{2}$ per cent less than that for the previous year, and about 31·6 per cent less than that for the year 1913. On the other hand, the production of lignite for 1922 was 137,207,000 tons, being 11·4 per cent more than that for 1921, and about 57·5 per cent more than that for 1913. These figures confirm those given by the United States Geological Survey.

LIGNITE, SHALE AND PEAT

Other potential sources of the world's fuel supplies are the brown coals, lignites, shale and peat. Extensive beds of brown coal occur in Germany, Central Europe, Victoria and Canada, but only to a very limited extent in England. In Germany the use of brown coal has been greatly developed in recent years. Since lignite, where available, is at the present time the cheapest source of thermal units, Germany's example is being followed by other countries. The importance which brown coal is likely to attain in future has been very clearly shown by Sir George Beilby,[1] and need not be recapitulated.

In the British Isles the second in order of importance as regards our fuel supplies are the oil shale deposits and the peat bogs. At present no commercial method has been developed to deal with the large percentage of sulphur unfortunately associated with some of the larger shale deposits which so seriously diminishes the usefulness and value of the oil products. In America and other countries, however, the problems connected with the utilisation of the vast deposits of oil shale are in some respects different to those in Great Britain. Neither in Great Britain nor abroad however, have these problems yet been satisfactorily solved.

The problems connected with the winning and drying of peat have also received much attention, but those arising from work on a large commercial scale are stil unsolved. Peat blocks lend themselves admirably to carbonisation at low tempera tures, but it is impossible for developments to occur in this direction until the prior problems of winning and drying have been satisfactorily dealt with. It i clear, for example, that before peat can be said to possess any economic value

[1] Beilby, Sir George. *Fuel Problems of the Future*. Inst. of Civil Engineers : James Forre Lecture, 1921.

machinery must be employed on a large scale for its winning and preparation. The drawback to the development and use of machinery for peat production is that, so far, the cost of production is more than the commercial value of the product, at least in localities where coal is within reasonable distance.

OIL

Of the world's alternative sources of fuel, oil is at present next in importance to coal. Of the output for 1920 of about 97 million tons of oil, the United States produced 64·8 per cent, and Mexico 23·3 per cent. The supply of natural oil in this country is practically negligible, although in times of peace, at all events, British interests in Eastern oil fields mitigate this disadvantage to some extent.

Estimates of the oil left in the oil fields of the United States indicate that they will last barely twenty years at the present rate of consumption. Probably, however, the production will be spread over a much longer period, though in later years it may be at a declining rate. There is therefore serious, though not immediate, concern for the future of the oil supplies from the United States oil wells. It is estimated that there still remain enormous reserves in Mexico and other countries ; though in some quarters the fear is expressed that Mexico's present phenomenal production may not much longer be continued.

The potential oil supplies to be obtained by the treatment of coal at low temperatures will be discussed in this volume, though no attempt will be made to consider the broader problem, viz. the heat treatment of all carbonaceous materials. Enough is known from laboratory experiments to justify the hope that, from these sources alone, it may be possible to draw a plentiful supply of oil to meet the world's needs for many centuries to come. Although full-scale experiments have not been equally successful, it is believed that the knowledge and experience gained during recent years will shortly make it possible to establish an industry on sound commercial lines whereby both oil and smokeless fuel supplies will be obtained in bulk, either from material which is at present consumed in a raw state, or, in a more limited number of cases, from the so-called " waste fuels."

ALCOHOL

As an alternative motor fuel to oil, alcohol may become of increasing importance. Crude oil has, however, an advantage over alcohol, inasmuch as it yields both lubricating oil and fuel oil. Alcohol, even if produced at a cheap enough rate, cannot therefore wholly supersede crude oil for industrial purposes. The problems connected with the available sources of supply and the manufacture of alcohol, its suitability for use in internal combustion engines and its denaturing for industrial

purposes, are discussed in the *Memoranda on Fuel for Motor Transport*,[1] published by the Fuel Research Board in July, 1920, and December, 1921. The investigations as to the possibilities of producing alcohol within the British Empire are still in progress.

THE UTILISATION OF THE HEAT OF COAL

The consideration of the economical use of our fuel supplies leads to a discussion of the third statement of the fuel problem, i.e. the methods whereby householders and manufacturers may obtain heat and power in the most convenient form for their own immediate needs. Fuel is not an end in itself, but merely a means to an end. The householder requires heat for the preparation of his food, and for the maintenance of those equable conditions of his dwelling, upon which the health and comfort of the inmates depend. To a minor extent he would often like also a small amount of heat energy as power, in order to be able to instal labour-saving devices in the home. On the other hand, the manufacturer only requires heat to a minor degree for the maintenance of an equable temperature in his factories. To a much greater extent he desires to have heat energy to supply his motive power and to drive his machines, or to initiate or produce some chemical change in the raw materials of his manufacturing processes. Occasionally he may wish to store potential energy which can be conveniently utilised at another spot, as, for example, by the pumping of water or by the manufacture of calcium carbide.

In the great majority of cases in which heat is utilised, however, the process eventually involves the complete loss to mankind of all the heat supplied. For example, in the case of a steamer crossing the ocean, fuel is burnt and transformed into work through the media of the boilers and engines. This work is used to drive the ship against the resistance of wind and water. In the process eddy currents and waves are set up, and when these finally die away the whole of the heat from the fuel has gone to raise infinitesimally the temperature of the surrounding sea and air.

Fuel is the chief, although not the only source of the energy of our modern industrial life. For convenience to householder and manufacturer, it may be desirable to transform the heat energy of coal into other forms of potential energy, as, for example, into gas and electricity for heat, power and light, etc. In any case, both the acquirement of the fuel and the transformation of its heat energy into the desired form are accompanied by losses which are occasionally many times that of the heat finally stored or utilised. The third aspect of the fuel problem is therefore concerned with methods of obtaining energy in some particular form at a definite spot with the least amount of loss.

[1] Fuel Research Board. *Interim Memorandum on Fuel for Motor Transport.*
    Fuel Research Board. *Second Memorandum on Fuel for Motor Transport.* (H.M. Stationery Office.)

AVAILABILITY OF HEAT

Sir George Beilby has used the term "availability of heat" in connexion with this aspect of the question. One fuel is then said to be of higher availability than another for a particular process when its heat can be used more conveniently or efficiently for that purpose. In all processes of fuel production and preparation it is desired to increase the availability of the heat, and thereby increase also the value of the fuel for special purposes. Coal in its native state underground cannot be used in industry until it has been won and raised to the surface. The processes of winning and raising increase, therefore, the heat availability. The coal may then be washed. Washing may make possible the more efficient use of the potential heat of the coal, and so may increase the availability of the heat energy of the fuel, which is then further increased (from the consumer's point of view) by its transportation to the user by rail or sea. The object of submitting coal to one of the processes of carbonisation is to raise further the availability of the heat units. In the form of gas or oil these units have a much greater degree of availability, rendering them more valuable to the fuel consumer, and enabling them to command a higher price than an equal number of heat units in that fuel in any of its original states.

THE INCREASE IN HEAT AVAILABILITY

All processes for raising the availability of heat are accompanied by losses. In coal washing, for example, a proportion of the "fines" is carried away with the ash in the slurry. In gasification, heat is lost in radiation and in the condensing plant. These processes also involve monetary expenditure in the form of labour charges and profit. Since the resulting fuel must bear all these costs, the problem becomes that of deciding whether the increased availability of the heat units in the finally produced fuel is worth the increased cost.

It will always be found that the actual heat employed in the utilisation process becomes more expensive as the availability of the fuel is increased, since each of the final heat units must be debited with its share of the British Thermal Units lost in the production of the final fuel. In other words, the over-all thermal efficiency of the process of obtaining and preparing the fuel is decreased as the availability of the heat is increased. It becomes necessary to judge in each case of commercial heat utilisation whether it is economically sound to use a large amount of heat from cheap fuel which cannot be utilised with high efficiency, or, on the other hand, to use a smaller amount of heat from a high grade fuel which can be used with high efficiency and great convenience. The prices commanded by a therm (viz. 100,000 British Thermal Units) in various forms of fuel differ according to the relative costs of the fuels at any time or place, but for convenience of reference are

given between wide limits in Table III. These figures are in themselves a measure of the value of the higher availability of special fuels. It may be said, therefore, that the aspect of the fuel problem thus presented is that of raising the availability of the fuel at the least possible cost.

TABLE III

Cost of One Therm in Various Forms of Fuel

	Pence.
Coke at 26/– to 40/– per ton, say 8 lb. per therm	1·1 to 1·7
Coal at 30/– to 50/– per ton, say 7½ lb. per therm	1·2 to 2·0
Fuel Oil at 5d. to 6d. per gallon, say 6 lb. per therm . . .	3·0 to 3·8
Gas at 9d. to 11d. per therm	9·0 to 11·0
Petrol at 1/4 to 2/6 per gallon, say 5 lb. per therm . . .	11·0 to 20·6
Electricity, 29 units at 1½d. to 3d.	43·5 to 87·0
Alcohol at 4/– to 5/– per gallon, say 8·3 lb. per therm . . .	49·6 to 62

APPLICATION TO LOW TEMPERATURE CARBONISATION

It will be seen in a later chapter that this aspect of the fuel problem is of particular importance to those interested in the development of the process of low temperature carbonisation. The availability of heat in coal is undoubtedly increased by carbonisation at a low temperature. The immediate problem to be solved by advocates of low temperature carbonisation is to make the cost of treatment of the original fuel sufficiently low to attract consumers generally, or for any particular industry, by the extra convenience and heat availability of the resulting fuels.

Although much might be written about the desirability and necessity of the utilisation of the so-called " waste fuels " and of the inferior fuels, such as shale or peat, it must be recognised that, in many cases, it is at present commercially unsound to attempt to increase their heat availability. There must always be a certain amount of waste fuel, no matter how scarce or expensive high-class fuel may become. It is only as the higher grades of fuel increase in price that increasing supplies of the lower grades of fuel become economically of importance. Examination will be made in later chapters of the claims made by some inventors of processes of low temperature carbonisation to utilise the fuels which are, under present industrial conditions, only potential sources of heat and power. It may safely be assumed that, even if certain of these processes are not commercially sound at the present moment, the changing conditions due to the undoubted gradual rise in the costs of the higher grade fuels may eventually enable some of the present processes, or improvements on the present processes, to become of commercial importance.

CHAPTER IV

NOTES ON THE CONSTITUTION OF COAL

INTRODUCTORY

ALTHOUGH coal is of very common occurrence and has been utilised for so long as a source of heat, light and power, it is undoubtedly true that the chemical and physical study of this important mineral has been much neglected. It is only within the last few years that the number of research workers who have been investigating different aspects of the constitution of coal has been in any degree commensurate with the importance of the subject. The research has also suffered by having taken the form of isolated experiments ; and only lately has there been any tendency for investigators to make a combined attack upon it. The reasons for the distinctive differences between various types of coal have not yet been completely placed upon a rational basis, nor is it possible to enunciate general laws regarding coal which are strictly true. A more intimate knowledge of the constitution, as distinct from the composition, of coal would undoubtedly be of great value to the technologist, and might assist in the solution of some of the problems relating to carbonisation.

DEFINITION OF COAL

It is very difficult to suggest a definition of coal which will be at once scientifically correct and of general acceptance. Stopes and Wheeler[1] define coal as follows :—

> " Ordinary coal is a compact stratified mass of mummified plants (which have in part suffered arrested decay to varying degrees of completeness) free from all save a very low percentage of other matter."

Strictly speaking, this definition is applicable to bituminous and anthracitic coals only, and does not cover the wider range of all carbonaceous material. Impure deposits, due to the plant substance being insufficiently free from inorganic matter, may grade into shales and other carbonaceous materials. On the other hand, to quote Lewes :[2]—

> " The most satisfactory view to take of the composition of coal is that it is an agglomerate of the solid degradation products of vegetable decay, together with such of the original bodies as have resisted to a greater extent the action to which the material has been subjected."

[1] Dept. of Scientific and Industrial Research. *Monograph on the Constitution of Coal.* Stopes. Marie C., D.Sc., Ph.D.: Wheeler. R. V., D.Sc. (H.M. Stationery Office.)
[2] Lewes, Vivian. *The Carbonisation of Coal.* (Benn Bros.)

39

THE ORIGIN OF COAL

Whatever may have been the mode of accumulation of coal, and the views of opposing theorists are not yet fully reconciled, it is at least universally recognised that coal is of plant origin. Notwithstanding all the internal evidence in its favour, it may nevertheless appear somewhat speculative to the lay mind that the vast deposits of coal should have had their origin in vegetable matter and plant life. The specific gravity of coal is of the order of 1.2, so that the volume of coal is only about one-tenth to one-twelfth of the volume of the original vegetable tissues. A seam of coal 10 feet thick is therefore the deposit from vegetation approximately 120 feet thick. Even when an unlimited geological time is assumed for its formation, an accumulation of this depth appears somewhat incredible, and the problem of accounting satisfactorily for an occurrence of this nature is profound. Even the parallel of the deposits possible from the rich tropical growths of the present day does not carry with it full conviction. In point of fact, however, the theory of the origin of coal should not be considered in the light of present day conditions. It must be borne in mind that physiographical conditions were radically different in the days of the luxuriant plant growths of the carbonaceous period to those of to-day. Excesses of atmospheric carbon-dioxide and water vapour, together with the heat from the cooling earth's crust, stimulated the growth of vegetable tissues. Atmospheric oxygen was just beginning to accumulate ; and decay, which is but the slow combustion of matter, did not operate as it does to-day. The decay which did occur must have been largely at the expense of the oxygen in the vegetable tissues ; that is to say, checked decay, as it is called, must have been operative. The later effects of pressure of an incalculable amount must be added to the former factors. Although these do not prove incontestably that the primeval forests and swamps have been transformed into the carboniferous measures, the internal evidence of the formation of coal from vegetable matter is additionally very strong. The results of microscopical examination, supported as they are by the impression of plant life found in coal, render it difficult to advance sufficiently strong arguments to refute this theory. There still remains much, however, that is uncertain regarding the earlier history of coal ; and in particular there are two problems on which there is not yet a consensus of opinion, viz. :—

(i) The mode of accumulation of coal, and

(ii) The mode of transformation of the vegetable matter into coal.

THE MODE OF ACCUMULATION OF COAL

Stopes and Wheeler[1] summarise very completely the leading modes of accumulation of coal-forming matter in four groups, viz. :—

(a) In sea water.

(b) In brackish water.

(c) In fresh water.

(d) On land.

There are two methods of accounting for the accumulation of our coal deposits under the above heads, viz. the *in situ* theory and the *drift* theory. In the first case it is assumed that the vegetable remains have not moved from their place of growth, and in the second that they have travelled from a distance by estuary and river action. The *in situ* theory is strengthened by the presence of stigmaria or fossil roots in the underclay of certain seams ; but the *drift* theory helps to account for the very thick coal seams which have been found. The impurities found in coal may be divided into two groups, viz. those deposited during the formation of the coal, consisting largely of silica and alumina, and those deposited subsequently, chiefly by percolation through the cleavage planes, e.g. calcium carbonate. For a correct study of the coal at any seam it is obviously of some importance that there should be some knowledge of its mode of accumulation ; but local experience is required in the majority of cases to decide whether the coal was formed of *in situ* or *drift* material, or perhaps both.

THE MODE OF TRANSFORMATION OF COAL

The theory of the origin of coal is supported by the occurrence throughout the world of carbonaceous material graded in all stages from the plant life of to-day to peat, from peat to lignite, from lignite to bituminous coal, and on to anthracitic coal. The theory has been advanced that, under the combined influences of pressure and temperature, and over vast geological epochs, these various gradations mark the mode of transformation of the coal of to-day from the vegetable growths of primeval times. While peat, for example, represents a condition of vegetable decay, intermediate between vegetable matter and the tertiary coals, it does not necessarily follow that vegetable matter on decay must pass through the peat stage before its metamorphosis into coal. It may be, therefore, that, as suggested by the metamorphic theory, the vegetable matter first underwent a rapid change due to the bacterial decomposition of its cellulose, and that a later slow alteration due to the combined effect of pressure and temperature [principally pressure, since the temperature cannot have approached 300° C.], has completed the transformation. It is obvious that, in the later stages of the coal-forming process at least, the effect

<hr>

[1] *Ibid.*, p. 39.

of temperature can only have been of subsidiary importance, since coal is readily decomposed at relatively low temperatures. The accumulated evidence appears to favour the metamorphic theory. In any case, the estimates made of the times taken for the accumulation and deposition of the original coal-forming debris and for its transformation into coal must be considered highly speculative.

THE DETERMINATION OF THE CHEMICAL CONSTITUTION OF COAL

Coal being composed of vegetable matter, it is to be expected that amongst its constituents will be :—

(a) Cellulosic or humic derivatives.
(b) Resinous derivatives, and
(c) Nitrogenous compounds.

The resinic constituents are believed to have a great influence on coke formation. There was formerly a disposition to classify the remaining constituents as "cellulosic," but it has been pointed out that cellulose itself is unstable under the conditions of peat formation. The chief differences in the properties of different coals may be said to be due to the varying proportions with which these derivatives are compounded. Three methods of research have been employed in determining the chemical constitution of coal, viz. :—

(i) The extraction of the constituents by means of solvents.
(ii) The action of reagents on the coal substance, and
(iii) Destructive distillation.

The first and most direct method of research has not yielded much information hitherto, apart from the use of alkaline liquids which may perhaps act as reagents. This is due to the fact that coal is of great insolubility and offers a strong resistance to the ordinary solvents. The second and third of the above methods are indirect, but nevertheless have proved useful in giving information of great value.

THE ACTION OF SOLVENTS UPON COAL

As already stated, the usefulness of this method of determining the chemical composition of coal is limited by the inertness of the coal substance. There is also a suspicion that some of the solvents employed modify very considerably the molecular structure of the compounds in the coal and act, indeed, as reagents rather than solvents. The solvents employed include :—

(i) Alcohol, ether, chloroform, acetone, aniline and petroleum.
(ii) Benzene and pentane.
(iii) Phenol.
(iv) Pyridine.

It is not necessary in this volume to study closely the results of the work of various experimenters with these solvents. It may be said, however, that a useful empirical separation of coal into three main groups of compounds has been effected by the use of pyridine and chloroform, thus :—

- (i) *Alpha* compounds, insoluble in pyridine.
  (ii) *Beta* compounds, soluble in pyridine, but insoluble in chloroform.
  (iii) *Gamma* compounds, soluble in pyridine, and soluble also in chloroform.

This separation may perhaps be seen more clearly from the following :—

BITUMINOUS COAL

treated with pyridine

Alpha Compounds   Extract A

Extract A
treated with chloroform

Beta Compounds   Gamma Compounds
(Insoluble)   (Extract B)
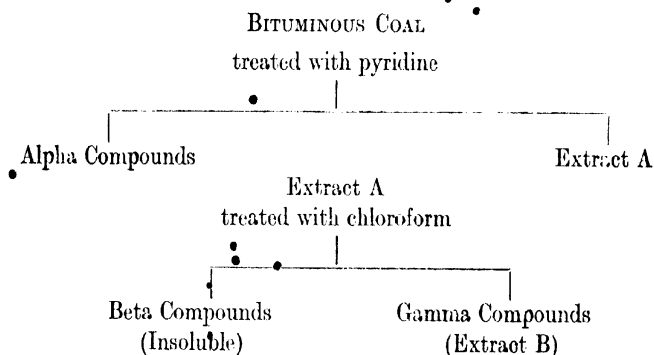
The alpha and beta compounds appear to be of the same type, and correspond in general characteristics to the derivatives of the celluloses (ligno-cellulose, etc.) in the coal, in contradistinction to the gamma compounds which appear to have concentrated in them the resinic derivatives. Until the composition of these groups of compounds has been more deffnitely established it is not desirable, however, to dogmatise about the latter classification. Jones and Wheeler[1] have found that the alpha and beta compounds yield on distillation very small percentages of liquid products, mainly phenols ; while about 40 per cent to 50 per cent of the gamma compounds distil below 400° C., the products being paraffins, olefins and naphthenes, but no phenols.

Stopes and Wheeler[2] summarise three main conclusions in their review of the experimental results obtained by solvents in groups (i) to (iv), viz. :—

(a) Evidence has been obtained of the presence in bituminous coals of small quantities (presumably but little altered) of resin extractable with ether.

[1] Jones and Wheeler. "The Composition of Coal." *Trans. Chem. Soc.*, 1915, **107**, 1318.
[2] Dept. of Scientific and Industrial Research. *Monograph on the Constitution of Coal.* Stopes, Marie C. D.Sc., Ph.D. ; Wheeler, R. V., D.Sc. (H.M. Stationery Office.)

(b) Free hydrocarbons exist in small quantities in many coals.

(c) Whatever be the exact nature of its action, pyridine affords a means of resolving many coals in such a manner as to render subsequent chemical examination less difficult.

Illingworth[1] makes the following generalisations :—

(i) Anthracitic coals are characterised by the general absence of beta and gamma compounds.

(ii) The properties of carbonaceous coals are due to the relatively high contents of alpha compounds compared with gamma compounds and to the absence of beta compounds.

(iii) Bituminous coals in general owe their most striking properties to the presence of gamma compounds ; while the beta compounds occur ir the greatest quantity in coals in which the $\frac{C}{H}$ ratio lies between 14 and 16.

ACTION OF REAGENTS ON COAL

Coal may be oxidised quickly by means of chemical reagents such as sulphuric and nitric acids, caustic alkalis, bromine and also ozone, or more slowly, as by the atmospheric oxygen, in the weathering of coal. It has been found[2] that coal boiled with a mixture of potassium chlorate and dilute hydrochloric acid yields chlorinated substances of complex composition entirely soluble in alkalies. Furthermore, many coals dissolve almost completely on evaporation with nitric acid. In most cases, however, the use of reagents yields products of great complexity, which throw little light on the nature of the original coal substance. Stopes and Wheeler thus summarise the position :—

" Considered by themselves, the facts elicited from the study of the action of reagents on coal are not very informing (we except, of course, the facts relative to the formation of ulmic compounds), nor are the prospects for future work in this direction very promising."

ULMINS

The action of potassium hydroxide must be considered separately, as the compounds removed from coal by its means are very important, and form the basis of the so-called " Ulmic Substances."

The Ulmins may be described as highly complex colloidal jellies which appear

[1] Illingworth, S. Roy, 'The Action of Solvents on Coal." *Fuel in Science and Practice*, June, 1922.

[2] Cross and Bevan. " On Pseudo Carbons." *Phil. Mag., & r.* 5, Vol. 13, pp. 325-8.

after the decay of the softer portions of plants. The ulmin compounds occur in wood decayed by " wet rot," to a lesser extent in peat and lignite, but very rarely in bituminous coal ; they have never been detected in anthracite. Thus the content of soluble ulmin compounds diminishes as the content of carbon increases. The formation or non-formation of ulmin compounds is indeed generally given as a method of distinguishing between a lignite and a bituminous coal. It is found, however, that a bituminous coal which has become oxidised, by weathering, for example, contains appreciable quantities of ulmin compounds, and that the quantity increases with the degree of oxidation. The presence of ulmin compounds detracts from the coking quality of a coal. A very interesting field for research is therefore opened up by a study of the ulmin compounds in carbonaceous material, and further information on the constitution of these compounds in coal would be of great value in determining the constitution of coal.

ATMOSPHERIC OXYGEN AND COAL

The most important reagent in the oxidation of coal is atmospheric oxygen. The spontaneous combustion and the weathering of coal, the reduction or prevention of which is of so much industrial consequence, are due to the slow absorption and combination of the oxygen of the air with the coal even at ordinary temperatures. It is not surprising, therefore, that considerable research work should have been done on this phenomenon. The weathering of coal results in the deterioration of its qualities, particularly as regards calorific value and coking properties. The weathering process begins with the absorption of oxygen, which unites chemically with the coal substance and generates heat. Should this heat not be dissipated, the temperature of the mass of coal slowly rises, due to its poor conductivity of heat, until the point of ignition is reached. The drop in calorific value is due to the oxidation of the carbon and hydrogen in the coal to carbon-dioxide and water respectively. Important factors in the spontaneous heating of coal are (a) the freshness of the broken coal surface—for the first few days or weeks the freshly broken coal surface is much more active in combining with oxygen than it is later ; (b) the temperature of the coal on storage—coal stored during the hot months of summer is more liable to spontaneous combustion than coal stored during winter ; and (c) the fineness of the coal—the surface of the coal is greatly increased as the size of the individual particles is diminished, and there are strong grounds for belief that the spontaneous heating of coal is a surface phenomenon.

Important researches, in which the results have been directly applied for industrial purposes, have been carried out by Sir Richard Threlfall on the spontaneous heating of coal,[1] particularly during shipment, and by Dr. J. S. Haldane on the

[1] Threlfall, R. " Spontaneous Heating of Coal." *Journ. Soc. Chem. Ind.* 1909, 28, 759.

spontaneous firing of coal in mines.[1] The former researches were carried out in connexion with the investigation of the New South Wales Royal Commission between 1896 and 1908, and the latter by the staff of the Doncaster Coal Owners' Research Laboratory.

The effect of the oxidation of coal on its coking properties will be discussed in a later section.

THE DESTRUCTIVE DISTILLATION OF COAL

Our chief knowledge of the constitution of coal has probably been obtained by the indirect method of destructive distillation, comparative results being obtained by the subjection of coal-forming materials and typical coals to the same process of destructive distillation. It is unnecessary to describe fully all the experimental work which has been carried out under this heading, some of which must be discussed in Chapter VI. The results of some of the researches have, however, some bearing upon the chemical constitution of coal, and it is desirable to summarise them in so far as they affect this aspect of the problem.

The following general conclusions of Burgess and Wheeler[2] are largely confirmed by the work of Porter and Ovitz,[3] and that of Vignon[4] :—

(i) There is for all coals a well-defined decomposition point, at a temperature lying between 700° C. and 800° C., which corresponds with a marked increase in the quantity of hydrogen evolved.

(ii) The evolution of hydrocarbon of the paraffin series practically ceases at temperatures above 700° C.

(iii) Ethane, propane, butane and higher members of the paraffin series form a large percentage of the gases evolved at temperatures below 450° C.

As a result of these observations, Burgess and Wheeler presumed that coal is composed of two compounds which differ in their tendency to decompose. The more unstable yields the paraffin hydrocarbons and no hydrogen ; the more stable decomposes rapidly between 700° C. and 800° C., yielding hydrogen as its chief gaseous product. They were further of opinion that the chief differences in the properties of coals are due to the proportions in which these compounds are blended.

Clark and Wheeler[5] gave additional evidence of the existence of these compounds, and labelled the paraffin-yielding compounds "resinic" and the hydrogen-

[1] Haldane, J. S. "Spontaneous Firing of Coal in Mines." *Proc. Inst. Min. Eng.*, 1917, 53, 194.

[2] Burgess and Wheeler. "The Volatile Constituents of Coal." *Trans. Chem. Soc.*, 1910, 97, 1917 ; 1911, 99, 649.

[3] Porter and Ovitz. "The Volatile Matter of Coal." *Bulletin* 79, U.S.A. Bureau of Mines.

[4] Vignon, L. "Distillation fractionée de la Houille" *Comptes Rendus*, 1912, 155, 1514–17.

[5] Clark and Wheeler. "The Volatile Constituents of Coal," Part III. *Trans. Chem. Soc.*, 1913, 103, 1704.

yielding "cellulosic" (see, however, page 42). Although it may be taken as confirmed that the evolution of hydrogen increases rapidly at a temperature between 700° C. and 800° C., it is not yet generally agreed that this phenomenon is due to the decomposition of a constituent of coal which is stable up to that temperature.

THE LIQUEFACTION OF COAL

Very interesting work on this subject has been done in Germany, under the direction of F. Fischer, at the Coal Research Institute at Mannheim. Since only about 6 per cent of the weight of the original coal is recovered as tar by the ordinary gasification processes, attempts have been made to obtain liquid fuel from solid fuel by means other than carbonisation. The action of ozone has been investigated, and other processes in relation to the oxidation and hydrogenation of coal under pressure. Research with ozone has been discontinued, partly because of the expense, and partly because the same results may be obtained by pressure-oxidation methods. In the latter, finely ground coal is mixed with a solution of sodium carbonate, after which it is treated with compressed air in steel containers at a temperature of 170° C. to 200° C. When subjected to this process, coal yields about 40 per cent of soluble organic compounds. The properties of these compounds have not as yet been closely determined. In the hydrogenation system of Bergius the coal is treated with hydrogen at about 400° C. under a pressure of 150 atmospheres in special steel cylinders. It is stated that about 80 per cent to 90 per cent of the original dry weight of certain coals has been in this way turned into an oil which shows the characteristics partly of phenolic and partly of the petroleum hydrocarbons.

As a result of their experimental investigations, Fischer and Schrader[1] consider that the main classes of compounds that have produced coal are :—

    (a) Cellulose, which is, however, destroyed during bacterial fermentation.
    (b) Lignin,[2] which becomes ulmins, and   .
    (c) Resin, which becomes "bitumen."

They contend that the greater proportion (if not all) of the cellulose disappears in the course of coal-formation through bacterial action, and that coal is composed chiefly of the ulmins produced by the transformation of lignone, though it may also be derived in part from the concentrated resins of the plants. According to this theory, which is not fully established, bituminous coal must be regarded as consisting mainly of (b) and (c) above. It should be noted that Fischer and Schrader differ from Wheeler in believing that the ulmic portion of coal is derived solely

---

[1] Fischer and Schrader. "Origin and Chemical Structure of Coal." *Fuel in Science and Practice*, July, 1922.

[2] It is perhaps preferable to use the term lignone, and this term is used hereafter in this volume.

from lignone (the lignone portion of ligno-cellulose), and not from the celluloses, and would prefer in Wheeler's writings to substitute the words lignone derivatives for cellulosic derivatives.

THE MICROSCOPICAL EXAMINATION OF COAL.

This consideration of the constitution of coal would be incomplete without some reference to the contribution afforded by macroscopical and microscopical evidence. One of the most interesting developments of recent years is the description by Dr. Marie Stopes of the four visible ingredients in banded bituminous coal. She points out that four distinctive and visibly differing portions are recognisable in the mass of a bituminous coal, and that these four portions " react so differently to certain simple chemical materials as to indicate that their chemical molecules should be substantially different from each other." To these she has given the following names :—

Fusain, equivalent to the so-called " mother of coal."
Durain, equivalent to the " dull " coal of various authors.
Clarain, and
Vitrain.

The two latter have been referred to indiscriminately by different authors as " bright coal."

Considerable experience is, of course, necessary before some of the constituents present in a block of coal can be identified with certainty. They occur frequently in thin bands, and tend in certain cases to merge ore into the other, e.g. it is sometimes difficult to distinguish, without the help of the microscope, between clarain and alternate thin bands of durain and vitrain. Stopes,[1] indeed, insists upon the fact that these portions—except possibly vitrain—are neither homogeneous nor chemical molecular units, and that clarain may eventually prove to be vitrain which contains plant remains. Mr. J. Lomax has done much work with the object of correlating the known general properties of the constituents with the commercial properties of typical coals. An interesting description of the methods employed by Mr. Lomax for observations and researches on the microstructure of coal is given in Fuel in Science and Practice, May, 1922.[2]

Fusain, according to Stopes, occurs as wedges roughly parallel to the bedding plane. It is very powdery, and, according to Sinnatt,[3] contributes greatly to the

---

[1] Stopes, Marie C. " On the Four Visible Ingredients in Banded Bituminous Coal." Proc. Roy. Soc. B., 90, 1919.

[2] Lomax, J. "The Preparation of Transparent Sections of Coal." Fuel in Science and Practice, May, 1922.

[3] Sinnatt, F. S., and Slater. "Propagation of a Zone of Combustion in Powdered Coal." Fuel in Science and Practice, Aug., 1923.
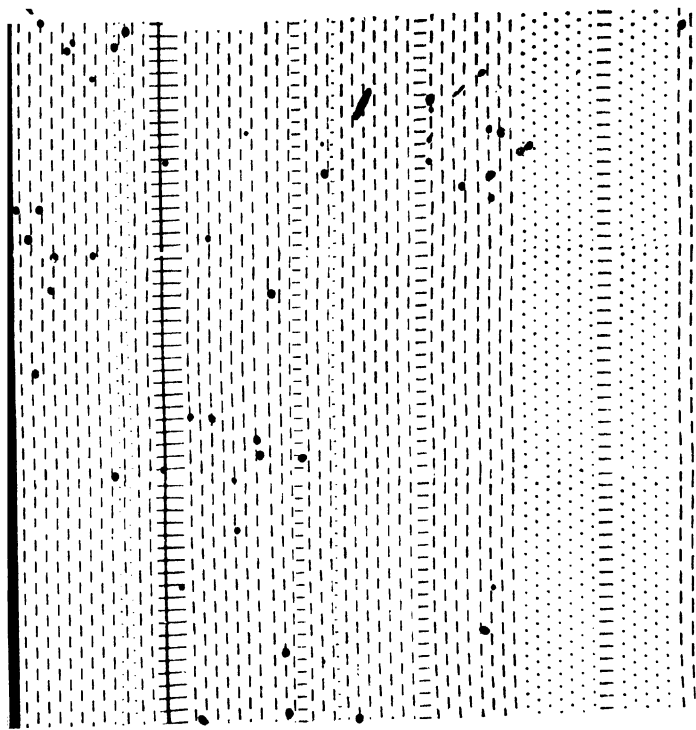
Fig. 1.—Coursing of Four Constituents in the Arley Coal.

dust which is so dangerous a factor in certain mines. The layers in which it occurs are utilised as lines of weakness by the miner in getting the coal. Examined microscopically in thin sections it is opaque and black, and often shows the vascular structure of the wood. In the coking test the residue appears as a non-coherent powder. In consequence dust from coal washeries may not possess a high coking power, despite the properties of the coal.

Durain, or spore coal, often referred to as dull coal, is hard and dull in appearance. It does not usually present a large surface of contact to adjacent constituents of other types. According to Stopes, " the purest durain tends to have ravelled edges." When examined in thin sections, it is seen to contain a large number of
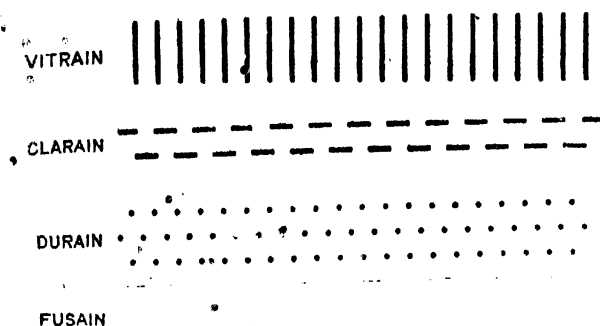
FIG. 2.—CONVENTIONAL NOTATION FOR COURSING DIAGRAM.

macrospores and microspores on a background of opaque granules. Sinnatt states that in certain special coals durain is practically representative of the whole of the seam, although in many cases it does not preponderate. Lessing[1] finds that on coking, durain undergoes practically no change in form, and that in general it must be regarded as almost devoid of coking value.

Clarain has a well-marked glassy appearance with a distinctly laminated structure. Microscopically, it has a clear appearance, though heterogeneous when compared with durain, and presents bands of leaf cuticle, stem tissues and resin bodies which are translucent with some spore exines. It is especially rich in plant remains, and on coking presents well-marked cohering properties.

Vitrain occurs in bands up to some half inch in thickness, is usually sharply defined, and is quite distinct from the neighbouring layers of other constituents. It does not present any structure, and tends to break in conchoidal form.

[1] Lessing, R. " The Behaviour of the Constituents of Banded Bituminous Coal on Coking." *Trans. Chem. Soc.*, 117, 1920.

Microscopically it is translucent with a uniform texture, and in colour a pale gold to red-brown, according to the thickness of the section. Stopes likens it to a hardened glue or jelly. Lessing has shown that vitrain is comparatively highly coking.

Although some use has been made of the microscopical examination of coals for certain commercial purposes, it cannot be said that up to the present there is any very definite correlation of the constitution of a seam to the use for which the coal may be suitable. Active work is, however, proceeding in this direction. Sinnatt[1] has recently plotted very carefully the vertical section of several seams, and has prepared a diagram showing the coursing of the four constituents in the Arley coal, which is reproduced with his permission in Fig. 1. It should be stated that, in this figure, the conventions given in Fig. 2 have been adopted by Sinnatt to indicate the four ingredients. It was at first thought that they could be indicated most conveniently on a drawing by giving them different colours, but this method has been rejected except for exhibition purposes.

TABLE IV

PERCENTAGES OF FOUR VISIBLE INGREDIENTS IN CERTAIN SEAMS

Name of Seam.	Vitrain.	Clarain.	Durain.	Fusain.	Foreign Matter.
Arley	14·6	65·9	18·1	1·4	—
Wigan 5 ft.	9·8	53·1	35·2	1·9	—
Trencherbone	7·0	70·0	21·2	1·8	—
,, Tops	7·2	55·8	35·5	1·5	—
,, Bottoms	6·8	85·6	5·6	2·0	—
Wigan 4 ft.	25·1	51·9	20·2	1·0	1·8
Arley	21·3	65·4	12·8	0·5	—
Queen Mine	1·3	96·7	Nil.	Nil.	—
Bickershaw Yard	Nil.	Nil.	100	Nil.	—

The percentages of the four ingredients in Arley coal and other seams are given in Table IV. Specimens of the ingredients were separated from four of the seams, and the results of the analysis of the material are given in Table V. The analysis of each ingredient in the Thick Seam, Hamstead Colliery, is given in Table VI.

[1] Sinnatt, F. S. "A Method of representing the Structure of Coal Seams." *Manch. Geol. and Min. Soc.*, June, 1922.

TABLE V

ANALYSES OF EACH INGREDIENT IN CERTAIN SEAMS

ARLEY MINE

	Moisture.	Ash.	Volatile Matter.	Volatile Matter less Moisture.	Coke.	Fixed Carbon.
	%	%	%	%	%	%
Vitrain	1·15	0·87	33·9	32·75	66·1	65·24
Clarain	1·04	1·15	33·0	31·96	67·0	65·85
Durain	1·06	2·50	30·4	29·34	69·6	67·1
Fusain	1·40	4·72	13·2	11·8	86·8	82·08

TRENCHERBONE

Vitrain	1·47	1·54	41·2	39·73	58·8	56·24
Clarain	1·35	2·17	44·1	42·75	55·9	53·73
Durain	1·31	1·47	35·9	33·56	64·1	62·64

WIGAN 5 FT.

Vitrain	7·0	1·92	39·9	32·9	60·1	58·18
Clarain	5·92	2·79	39·8	33·88	60·2	57·41
Durain	3·58	4·65	37·9	34·32	62·1	57·45

WIGAN YARD

Vitrain	3·4	6·82	37·4	34·0	62·6	61·78
Clarain	3·2	3·26	41·4	38·2	58·6	55·34
Durain	2·1	12·90	28·8	26·7	71·2	58·30
Fusain	1·7	5·90	23·7	22·0	76·3	70·40

TABLE VI

ANALYSIS OF EACH INGREDIENT IN THICK SEAM, HAMSTEAD COLLIERY[1]

	Moisture.	Ash.	Volatile Matter in Ash-Tree dry Coal.
Vitrain	12·6	1·2	38·6
Clarain	10·2	1·45	40·8
Durain	6·5	3·6	39·4
Fusian	3·9	10·0	22·6

[1] Tideswell, F.V., and Wheeler, R.V. "A Chemical Investigation of Banded Bituminous Coal" Trans. Chem. Soc., 1919, 115, 630.

It must be admitted, however, that up to the present the microscope has been useful for comparative purposes only. Further work is needed before it will be possible to show definitely that, if the constituents are present in a certain proportion, the coal will be useful for any specific purpose. Typical samples of coal which have proved satisfactory, and of others which have not proved satisfactory, may be examined, and it may be found that coals with a similar appearance under the microscope behave similarly in practice. For commercial purposes, such as the selection of coal for a definite contract, the microscope may usefully be employed in eliminating unsuitable types; but with the present knowledge the final selection should preferably not be made until a trial under working conditions has been carried out.

CHAPTER V

THE CLASSIFICATION AND TESTING OF COAL

THE CLASSIFICATION OF COAL

IT is very difficult to classify the different varieties of carboniferous deposits as they exist to-day. Several methods have been suggested, but it cannot be said that any one is entirely satisfactory. Thus the classification may be made from the point of view of the geological age, the coking properties, the commercial application, the chemical composition, etc.; but as the characteristics in any one group tend to merge into those of another, the various schemes of classification are convenient rather than scientific. Six grades may be distinguished, viz. :—

(1) Peat.
(2) Lignites or Brown Coals.
(3) Cannel Coals.
(4) Torbanites.
(5) Bituminous Coals.
(6) Anthracites.

The difference in the chemical composition of carbonaceous materials is exemplified by the average figures given in the following table by Butterfield :—

TABLE VII

CHEMICAL COMPOSITION OF CARBONACEOUS MATERIALS

	Carbon.	Hydrogen.	Oxygen.	Nitrogen.	Sulphur.	Ash.
Cellulose	44·4	6·2	49·4	—	—	—
Dry Wood (average)	48·5	6·0	43·5	0·5	—	1·5
Dry Peat	58·0	6·3	30·8	0·9	trace	4·0
Lignite	67·0	5·1	19·5	1·1	1·0	6·3
Coal	77·0	5·0	7·0	1·5	1·5	8·0
Anthracite	90·0	2·5	2·5	0·5	0·5	4·0

It will be noted that the principal differences from cellulose to anthracite are in the gradual and almost total elimination of oxygen—and to a smaller extent of hydrogen also—and the corresponding increase in the carbon content.

GRÜNER'S CLASSIFICATION

It is not necessary to give in detail the various classifications of coal which have been advanced. One of the earliest, first suggested by Regnault and slightly

modified by Grüner, has been widely adopted on the Continent, and is given in Table VIII. This classification is based on dry coals which are ash-free and sulphur-free.

TABLE VIII

GRÜNER'S CLASSIFICATION OF COAL

Class.	Composition per cent.			Specific Gravity.	Nature of Coke.
	Carbon.	Hydrogen.	Oxygen.		
(1) Dry coals long flame	75–80	5·5–4·5	19·5–15·0	1·25	Powdery or slightly fritted.
(2) Fat coals long flame	80–85	5·8–5·0	14·2–10·0	1·28–1·30	Caked, but porous.
(3) Fat coals	84–89	5·0–5·5	11·0–5·5	1·30	Caked and fairly compact.
(4) Fat coals short flame	88–91	4·5–5·5	6·5–5·5	1·30–1·35	Compact.
(5) Lean coals anthracite	90–93	4·5–4·0	5·5–3·0	1·35–1·40	Lightly fritted or powdery.

SEYLER'S CLASSIFICATION

One of the best methods so far suggested for the purpose of the scientific classification of coal is that put forward by Mr. C. A. Seyler.[1] As the result of a detailed study of the coals of South Wales, which range in many varieties between bituminous and anthracitic coal, coals are divided according to their hydrogen content into five principal groups, each of which is further subdivided into other classes according to the carbon content. The chief classes of each group are shown in Table IX. This classification has been used by Drs Strahan and Pollard,[2] of the Geological Survey of England and Wales, in their publication upon the coals of South Wales.

[1] Seyler, C. A. "The Chemical Classification of Coal." Proc. S. Wales Inst. Eng., 1900, 21, 483–526.
[2] Strahan and Pollard. "The Coals of South Wales, with special reference to the Origin and Distribution of Anthracite." Ed. 2, Mem. Geol. Survey, Eng. and Wales, i., 91. (H.M. Stationery Office.)

TABLE IX

SEYLER'S CLASSIFICATION OF COAL

Hydrogen. Per cent.	Genus.	Carbon. Per cent.						
		Over 93·3	93·3 to 91·2	91·2 to 89·0	89·0 to 87·0	87·0 to 84·0	84·0 to 80·0	80·0 to 75·0
		Anthracitic.	Carbonaceous.	BITUMINOUS.			LIGNITOUS.	
				Meta.	Ortho.	Para.	Meta.	Ortho.
Over 5·8	Per-bituminous	—	—	Per-meta-bituminous	Per-ortho-bituminous	Per-para-bituminous	Per-lignitious	
5·0 to 5·8	Bituminous	—	Pseudo-bituminous	Meta-bituminous	Ortho-bituminous	Para-bituminous	Meta-lignitious	Ortho-lignitious
4·5 to 5·0	Semi-bituminous	—	Ortho-semi-bituminous	Sub-meta-bituminous	Sub-ortho-bituminous	Sub-para-bituminous	Sub-meta-lignitious	Sub-ortho-lignitious
4·0 to 4·5	Carbonaceous	Semi-anthracitic	Ortho-carbonaceous	Pseudo-carbonaceous (Sub-meta-bituminous)	Pseudo-carbonaceous (Sub-ortho-bituminous)	Pseudo-carbonaceous (Sub-para-bituminous)		
Under 4·0	Anthracitic	Ortho-anthracitic	Sub-carbonaceous	Pseudo-anthracitic (Sub-meta-bituminous)	Pseudo-anthracitic (Sub-ortho-bituminous)	Pseudo-anthracitic (Sub-para-bituminous)		

U.S.A. GEOLOGICAL SURVEY CLASSIFICATION

The United States Geological Survey[1] has adopted a classification somewhat similar to that proposed by Mr. Seyler, the basis being the ratio of the carbon-hydrogen content. This classification is given in Table X.

TABLE X

U.S.A. GEOLOGICAL SURVEY CLASSIFICATION OF COAL

Group.	Description.	Ratio : C.H.
A	Graphite	∞ to (?)
B ⎱ C ⎰	Anthracite	(?) to 30 (?) ⎱ (?) 30 to 26 (?) ⎰
D	Semi-anthracitic	(?) 26 to 23 (?)
E	Semi-bituminous	(?) 23 to 20
F ⎫ G ⎪ H ⎬ I ⎭	Bituminous	20 to 17 ⎫ 17 to 14·4 ⎪ 14·4 to 12·5 ⎬ 12·5 to 11·2 ⎭
J	Lignitic	11·2 to 9·3 (?)
K	Peat	(?) 9·3 to (?)
L	Wood	7·2

PARR'S CLASSIFICATION

A new classification of coal has been proposed by Mr. S. W. Parr,[2] of the University of Illinois, at the recent Annual Congress of the American Chemical Society. This classification is based upon the heating value per lb. of coal substance and the percentage of volatile matter contained in the coal. The unit heat value of the coal substance is determined from the formula :—

$$\frac{\text{Total B.Th.U.} - 5000\,S}{1 - (1.08\,A - 0.55\,S)}$$

where S = percentage of sulphur.
and A = percentage of ash.

This formula gives a true heat value for the coal substance itself without regard to the percentage content of the ash or sulphur of the coal.

This classification would group American coals as shown in Table XI.

[1] U.S.A. Geological Survey : Professional Paper No. 48. Report on Coal Testing Plant, 1906.
[2] Parr, S. W. "The Classification of Coal." *Journ. of Ind. and Eng. Chem.*, 1922, **14**, 919.

TABLE XI

PARR'S CLASSIFICATION OF COAL

Class.	Heating Value per lb. of Coal Substance.
1. Cellulose and Wood . . .	7500– 8500
2. Peat	7800–11500
3. Brown Lignite . . .	11500–12500
4. Black Lignite	12500–13500
5. Sub-bituminous . . .	13500–14000
6. Western Bituminous. (American) .	14000–15000
7. Eastern Bituminous. („) .	15000–16000
8. Anthracite	15500–16000

In conclusion, it should be added that, although each of the above classifications has doubtless its utility, it cannot yet be said that any classification has been evolved which enables the practical engineer to say definitely that coal of any particular type can be most economically applied for a particular purpose.

FURTHER CLASSIFICATIONS OF COAL FROM A CARBONISING STANDPOINT

Attempts have been made from time to time by various investigators of coking problems to take advantage of some of the information which has been obtained on the constitution of coal. In one interesting series of patent specifications[1] Illingworth describes how by subjecting coal to pre-heat treatment he is able to decompose some of the constituents which have already been referred to, and so to modify the raw material by adjusting the undecomposed portions to correspond to the proportions of similar constituents in a coal known to give the final product desired. Thus, in specification 164,104 (1921), referring to the production of metallurgical coke, he classifies coals as follows :—

Type I. In which the humic, cellulosic and resinic substances decompose at or below 300° C.

„ *II.* Decomposition at or below 350° C.

„ *III.* Decomposition at or below 400° C.

„ *IV.* Decomposition at or below 450° C.

Illingworth goes on to state that the best types of coking coal and also gas coals contain certain amounts of Types II, III and IV, but that the content of Type II is greater in a gas coal than in a good coking coal. He further states that

[1] Illingworth, S. Roy. Pat. Spec. No. 164,104 (1921), 175,838 (1922), 186,085 (1922), 187,328 (1922).

the more volatile gas coals contain considerable quantities of Type I. In his own words : " If, therefore, gas coals are heated out of contact with air to 350° C., coals of the Types I and II may be eliminated, leaving a residue containing Types III and IV, such residue being suitable for coke production."

In a similar way Illingworth describes in later patents the manner in which the properties of a coal which gave a soft semi-coke on direct carbonisation at 500° C. could be modified by pre-heat treatment so as to form a dense, hard, smokeless fuel at 480° C. containing some 10·3 per cent of volatile matter.

In the Report of the Fuel Research Board,[1] coals for the purposes of carbonisation are classed under two heads :—

(a) Those in which the resulting coke occupies a greater volume than the original coal, and

(b) Those in which it occupies a smaller volume.

For the present, and until our knowledge of the constituents of coal is greatly increased, this latter classification appears to meet all the requirements of low temperature carbonisation practice. By mixing the two types in correct proportions, a coke can be produced which meets all the requirements of strength and robustness to withstand the rough handling of transport and distribution. The proportions can be determined in a few hours by the method of low temperature assay which is briefly described on page 60.

TESTS ON COAL

Various tests are made on coals in order to obtain information upon their quality and to determine their suitability for various commercial purposes. Useful though this information may be, it is by no means a conclusive indication of the properties of the coal under test. Coal is a heterogeneous mixture of compounds with widely differing physical and chemical features, and it is not to be expected that the results of any simple test will reveal all the important characteristics of the coal. Indeed, any advance in the study of the potential behaviour of coal must be slow so long as coal is treated as a unit compound and not as a mixture of substances. In default of any more suitable test, it has been the practice to correlate the properties of coal with the results of the following tests amongst others :—

(i) Proximate analysis, i.e. the determination of the percentage by weight of the moisture, volatile matter, "fixed" carbon and ash.

(ii) Ultimate analysis, i.e. the determination of the percentage by weight of the elements, carbon, hydrogen, oxygen, nitrogen and sulphur.

(iii) Estimation of the calorific value.

(iv) Determination of the coking (caking) index.

[1] Fuel Research Board : Report for the Years 1920, 1921 ; Second Section : " Low Temperature Carbonisation." (H.M. Stationery Office)

The great difficulty in making estimates from the results of these tests is that the analytical processes themselves affect the coal, particularly in the determination of moisture and the volatile constituents. A certain amount of occluded gas, for example, may be obtained. The occluded gases may be determined by pumping off the gas liberated or by collecting the gas given off in a vacuum, either at ordinary temperatures or 100° C. It is found that nitrogen and oxygen generally come off first, followed more slowly by carbon dioxide and methane, but at ordinary temperatures such experiments will last for many days.

It is not necessary to describe the methods whereby these various determinations may be made, which must for the above reasons be considered largely conventional. Those employed in industrial laboratories may differ from those essential for scientific investigation, in which a greater degree of accuracy, and consequently

TABLE XII

TYPICAL DIFFERENCES BETWEEN PROXIMATE ANALYSES ON MOISTURE-FREE AND ASH-FREE BASES

	A	B	C	D	Moisture-Free. A	B	C	D	Moisture-Free and Ash-Free. A	B	C	D
Moisture	7·20	3·50	6·37	4·84	—	—	—	—	—	—	—	—
Volatile Matter	32·15	33·47	33·40	30·26	34·64	34·68	35·67	31·80	39·01	36·62	37·25	35·0
Carbon	50·25	57·93	56·26	56·19	54·15	60·03	60·09	59·04	60·99	63·38	62·75	64·9
Ash	10·40	5·10	3·97	8·71	11·21	5·29	4·24	9·16	—	—	—	—

more complicated apparatus and processes, may be necessary. · In passing it should be pointed out that it is of the utmost importance that the small quantity of coal used in the tests should have so far as possible the average quality of the bulk of coal under examination. The sampling of the coal, i.e. the selection of the small quantity of coal used in the tests, should therefore be effected in a standard manner, which also need not be described in this volume.

It will be noted that, in the proximate analysis of coal, the percentage content of moisture, volatile matter, "fixed" carbon and ash are stated. In order to give the full data, the moisture content should be determined:—

(a) On freshly mined coal.

(b) On air-dried coal after crushing.

The ash may be expressed either as the percentage content of original inorganic matter in the coal or as the residue left after complete combustion. There is a

great difference between these two results, and the second is that generally stated. Since the qualities of the two coals whose proximate analyses are known may be masked by the percentage content of moisture and ash, the proximate analysis is sometimes given in terms of moisture-free and ash-free coal. Typical differences arising in this way are shown in Table XII.

It may be added that it does not necessarily follow that the higher the coking index of a coal, the denser and harder will be the product of carbonisation. On the contrary, the density, toughness and crushing strength of cokes can often be increased by an addition of inert matter. This point will be discussed later.

LOW TEMPERATURE ASSAY

Until comparatively recently the development of a process for the carbonisation of coal at low temperatures has been greatly hampered by lack of knowledge of the quantity and quality of the resulting liquids and gases when carbonaceous materials are heated to different temperatures for definite periods. The ordinary laboratory methods of testing supply no direct information on these points, nor on the equally important questions of the rate of evolution of the gases, nor the effect of the temperature and the time of carbonisation on the quality of the resultant coke. The low temperature assay of Gray and King[1] provides a method which affords accurate information upon these points, and their apparatus is gradually being recognised as an essential adjunct to the equipment of those interested in the development of practical methods of low temperature carbonisation. Their apparatus, which was worked out at H.M. Fuel Research Station, is based on a method which had been used for many years in the Scottish shale oil industry, and consists of a glass or silica tube about 2 cm. in diameter and 30 cm. in length, closed at one end. At a distance of about 2 cm. from the open end a side tube, 1 cm. in diameter, is provided, which acts as an offtake pipe leading the gases into a condensing and scrubbing system.

The complete lay-out of the laboratory assay apparatus is shown in Fig. 3. B is the silica retort heated in the furnace A. The tube C acts as a condenser and is provided with an extension piece for the reception of the liquid products. The tube D is filled with glass beads drenched with sulphuric acid for the absorption of ammonia. The gas is collected either over a mixture of glycerine and water, a solution of magnesium chloride in water, or water saturated with the gas obtained in a previous experiment. A constant pressure in the gas holder is maintained by the ingenious apparatus shown at G, H, J, K. The gas entering the gas holder E

——— [1] Fuel Research Board. Tech. Paper No. 1 : " The Assay of Coal for Carbonisation Purposes : a new Laboratory Method." Thos. Gray, D.Sc., Ph.D., and Jas. G. King, F.I.C., A.R.T.C. (H.M. Stationery Office.)

displaces liquid which flows into G through a piece of india-rubber tubing. The overflow from G passes into the reservoir K, in which floats a counterpoise J. J and G are connected by a cord passing over the two pulleys shown. Thus, as the level in K rises, the receptacle G is lowered ; and if E and K have equal cross-sectional areas, a constant pressure is automatically maintained in the gas holder E. By an adjustment of the position of G at the beginning of the experiment any desired difference of level may be steadily maintained throughout the test.

The furnace A may be either gas fired or electrically heated, and heats uniformly
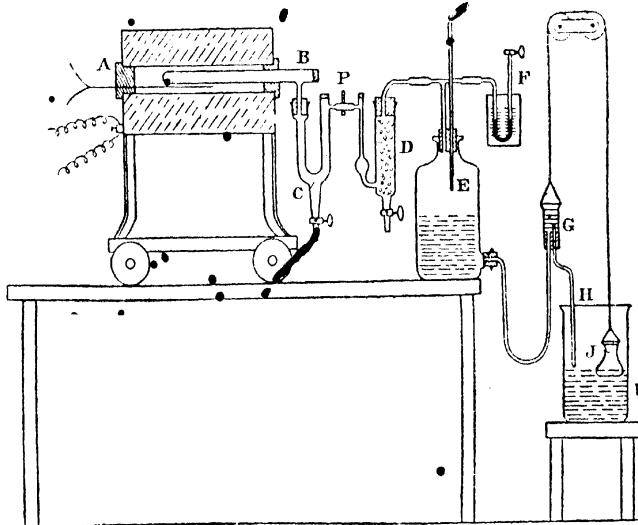
FIG. 3.—LOW TEMPERATURE ASSAY APPARATUS
(Fuel Research Board).

at least 6 inches of the tube B. The temperature is under control, and may be observed from time to time by means of a thermocouple in which the hot junction is in contact with the middle of the retort tube, as shown in the diagram.

This apparatus is now obtainable from at least two of the well-known firms of apparatus manufacturers, and has been recommended for the use of analysts and others by the Sampling and Analysis Committee of the Fuel Research Board.[1] It meets the long-felt want of an apparatus by which within a few hours accurate information may be obtained of the quality of a coal for carbonisation purposes, and from which the most desirable treatment for any coal or blend of coals can

[1] Fuel Research Board. Physical and Chemical Survey of the National Coal Resources, No. Interim Report on Methods of Analysis of Coal. (H.M. Stationery Office.)

TABLE XIII

Comparison of Yields obtained in Low Temperature Laboratory Assay (L) and Horizontal Retorts (H) expressed as parts per ton of Dry Coal.

	Langley Brights. Tupton Seam.	Dalton Main Hards. 1	Dalton Main Hards. 2	Mitchell Main Gas Nuts.	Ellistown Main Breeze.	60 per cent Mitchell Main, 40 per cent Ellistown Main.	Mean Ratio, Horizontals: Laboratory.
Coal (dry)	100·0	100·0	100·0	100·0	100·0	100·0	
Coke (cwts.)—							
L	14·66	14·83	15·38	15·38	14·61	15·11	
H	14·76	14·86	15·77	15·72	15·22	15·34	
Ratio H : L	1·01	1·00	1·02	1·02	1·04	1·02	1·02
Tar (gallons)—							
L	28·2	30·4	25·0	26·8	21·5	25·2	
H	16·5	17·0	14·0	16·25	12·9	15·55	
Ratio H : L	0·59	0·56	0·56	0·60	0·60	0·62	0·60
Liquor (nett gallons)—							
L	12·3	8·4	10·1	7·85	17·9	11·2	
H	17·15	11·55	11·7	6·95	17·1	14·5	
Ratio H : L	1·39	1·38	1·16	0·89	0·96	1·29	1·18
Ammonium Sulphate (lb.)—							Mean ratio with suitable scrubbing.
L	17·4	4·35	4·80	4·80	9·6	7·0	
H	—	3·6	4·18	2·73	8·45	6·1	
Ratio H : L	—	0·83	0·87	0·57	0·88	0·87	1·0
Gas (c. ft. at 30° and 60° F. satd.)—							
L	3770	3770	3370	3510	3450	3480	
H	3300	3770	3150	3480	3240	3210	
Ratio H : L	0·95	1·00	0·94	0·99	0·95	0·93	0·96

be deduced with the full confidence that, within certain limits, the results can also be obtained on a working scale. For a full description of the method of operation and of typical results obtained by the apparatus, reference should be made to the pamphlet already quoted.

CORRELATION OF ASSAY AND FULL-SCALE RESULTS

Further work on the correlation of the laboratory assay with the results obtained in retorts built upon a working scale is discussed in the Report of the Fuel Research Board for the years 1920–21, second section.[1] Table XIII gives a comparison of yields obtained in the low temperature assay and horizontal retorts expressed as weights per ton of dry coal carbonised. It will be seen that, in the cases of coke, liquor, ammonium sulphate and gas, the mean ratio is in the neighbourhood of unity; but that, in the horizontal retorts used at H.M. Fuel Research Station, the oil yield only averaged 60 per cent of that obtainable from the assay. It is, of course, not impossible that this ratio could be raised by modifications in this particular plant. There is undoubted evidence of cracking taking place, due to the fact that the vapour remains for some considerable time exposed to radiant heat, as well as being in contact with heated surfaces. While some improvement is possible, the low ratio of tar yield is typical of the losses which usually occur in a full-scale plant. This emphasises the necessity of a reliable conversion factor between the results of laboratory testing and those obtained in large-scale operations, and shows the danger of accepting the estimates of the yields of the highly priced products of carbonisation which are based solely on the results of ordinary laboratory testing.

[1] Fuel Research Board. Report for the Years 1920, 1921; Second Section: "Low Temperature Carbonisation." (H.M. Stationery Office.)

NOTES ON THE CHEMISTRY OF CARBONISATION PRODUCTS

INTRODUCTORY

By the distillation of coal a large number of chemical compounds is obtained in the gas, liquor and tar; some three or four hundred substances have been identified in the products of high temperature coal distillation, and doubtless many of these are to be found in the tar, etc., resulting from low temperature distillation. These substances, which do not occur as such in the coal, are formed either by the thermal decomposition of the coal substance (primary products), or by decomposition of the primary products, or by chemical interaction of the primary products with one another in the presence of incandescent coke. The substances formed from the primary products may be called secondary products. It is not possible in such a work as this to give a detailed account of the chemistry of the individual substances which have been identified among the products of low temperature coal distillation; for such an account reference must be made to the standard works on organic chemistry. In what follows it is assumed that the reader has a knowledge of the outlines of that science.

In this section it is proposed to give classified statements of those bodies which are indicated in the literature as having been identified, together with a few explanatory notes. It must be remembered that much of the work upon low temperature carbonisation tars is of very recent date, and, consequently, the results in some cases are still under discussion and may be subject to revision. Further, the lists given here by no means represent *all* the compounds present in low temperature tars; much investigation remains to be done, and it is to be expected that their number will steadily increase. Finally, from the complex mixtures encountered in these tars, composed as they are of many substances chemically akin to each other, it is very frequently a matter of extreme difficulty to separate pure substances. Progress towards a full knowledge of the constituents of low temperature tars is and will be, of necessity, slow.

CLASSIFICATION OF THE PRODUCTS OF LOW TEMPERATURE DISTILLATION

According to the elements they contain, the products of low temperature coal distillation may conveniently be classified as follows:—

> A. Hydrocarbons.
> B. Compounds containing oxygen.
> C. Compounds containing sulphur.
> D. Compounds containing chlorine.
> E. Compounds containing nitrogen.

some cases these main groups are capable of subdivision, as we shall see later in aling with them in the above order.

A. HYDROCARBONS

The hydrocarbons may be subdivided into the paraffins, ethylenes, cyclo-raffins, acetylenes and benzenes ; not included in these groups are a number of her hydrocarbons to which reference will be made.

1. PARAFFINS

· The following list gives the paraffin hydrocarbons which have been identified in w temperature coal distillation products :—

Paraffin Hydrocarbons.

Methane	CH_4	
Ethane	C_2H_6	Frequently mentioned in the literature of the subject.
Propane	C_3H_8	
Butanes	C_4H_{10}	

n-Pentane C_5H_{12} — Parr and Olin, " The Coking of Coal at Low Temperature," *Bulletin* 79, Eng. Expt. Stat., Univ. of Illinois.
Schütz, Buschmann and Wissebach, " Low Temperature Coal Tar and the Products of its Overheating." *Ber.*, 1923, **56**, 1091.

Methylbutane C_5H_{12} — Schütz, Buschmann and Wissebach, " Low Temperature Coal Tar and the Products of its Overheating." *Ber.*, 1923, **56**, 1091.

n-Hexane C_6H_{14} — Parr and Olin, and Schütz, Buschmann and Wissebach, *loc. cit.*

Methylpentane C_6H_{14} — Schütz, Buschmann and Wissebach, *loc. cit.*

n-Heptane C_7H_{16} — Parr and Olin, and Schütz, Buschmann and Wissebach, *loc. cit.*

n-Octane C_8H_{18} — Schütz, Buschmann and Wissebach, *loc. cit.*
Decane $C_{10}H_{22}$ — Fischer and Gluud. *Ber.*, 1919, **52**, 1053.
Solid paraffins $C_{19}H_{40}$ — Fischer and Gluud. *Ber.*, 1919, **52**, 1053.

$C_{23}H_{48}$ (M. Pt. 47·7°) — Fromm and Eckard, " Lignite Low Temperature Tar." *Ber.*, 1923, **56**, 948.

$C_{24}H_{50}$ (M. Pt. $51 \cdot 1°$) $\left\{ \begin{array}{l} \text{Fromm and Eckard, } loc.\ cit.;\ \text{Gluud,} \\ \quad Ges.\ Abhandlungen\ zur\ Kenntnis\ der \\ \quad Kohle,\ 1917,\ \mathbf{2},\ 301. \end{array} \right.$

$C_{26}H_{54}$ (M. Pt. $44°$)   Fromm and Eckard, loc. cit.

$C_{26}H_{54}$ or $C_{27}H_{56}$ $\left\{ \begin{array}{l} \text{Jones and Wheeler, "The Composition} \\ \quad \text{of Coal," } Trans.\ Chem.\ Soc.,\ 1914,\ \mathbf{105}, \\ \quad 140. \end{array} \right.$

$C_{28}H_{58}$ (M. Pt. $62°$)   Fromm and Eckard, loc. cit.

$C_{29}H_{60}$ Gluud, loc. cit.

$C_{32}H_{66}$ (M. Pt. $69 \cdot 6°$) Fromm and Eckard, loc. cit.

$C_{34}H_{70}$ (M. Pt. $73°$)   Fromm and Eckard, loc. cit.

As will be seen, this list contains a number of members of the homologous series of paraffins, the general formula of which is C_nH_{2n+2}. Structural isomerism commences at the butanes, of which there are two having the same empirical formula, C_4H_{10}, but different graphic formulæ; the two butanes thus have molecules of the same size, but the molecular architecture is different, and consequently the two substances are perfectly distinct. There are three isomeric pentanes, and, as we pass up the paraffin series, the number of possible isomerides having one particular empirical formula rapidly increases. Thus there are no less than 355 possible isomers having the formula $C_{12}H_{26}$. The higher members of the paraffin series (the solid paraffins, for instance), which have been identified in low temperature tars, in all probability consist of a number of isomerides which it would be extremely difficult, if not impossible, to separate from each other.

The paraffins found in low temperature tars exhibit that regular gradation in physical properties which is characteristic of an homologous series; there is, for instance, a steady rise in density up to a maximum of about $0 \cdot 78$ as the series is ascended.

Chemically, the paraffin hydrocarbons are characterised by their comparative inertness. They do not readily react with other substances, and such activity as they possess decreases with increasing molecular weight. This is partly due to the fact that they are stable, saturated compounds, and consequently can only form new derivations by the replacement of one or more of the hydrogen atoms in their molecules by other elements or radicles. Thus they are not absorbed by bromine or sulphuric acid; fuming nitric acid has little or no action on the lower members of the series, and other oxidising agents, such as chromic acid and potassium permanganate, have similarly little effect. Under suitable conditions chlorine and bromine interact with the paraffins, yielding halogen substitution derivatives, and the corresponding halogen hydracids are by-products.

2. ETHYLENES

The ethylenes or olefines are unsaturated hydrocarbons having the general formula C_nH_{2n}. Those which have so far been identified in the products of low temperature distillation are as follows :—

Ethylene		C_2H_4.	
Propylene		C_3H_6.	
Butylene	$\varDelta^{\alpha\beta}$	C_4H_8,	$(CH_2: CH \cdot CH_2 \cdot CH_3)$.
Butylene	$\varDelta^{\beta\gamma}$	C_4F_3,	$(CH_3 \cdot CH : CH \cdot CH_3)$.
Pentene (Amylene)	$\varDelta^{\alpha\beta}$	C_5H_{10},	$(CH_2 : CL \cdot CH_2 \cdot CH_2 \cdot CH_3)$.[1]

The above compounds are all members of the homologous ethylene series exhibiting the characteristic gradation of physical properties ; up to and including the butylenes the compounds are gases ; the amylenes are volatile liquids, and the boiling-points of the succeeding members rise until at octadecylene ($C_{18}H_{36}$) we reach a compound which is solid at ordinary temperatures (M. Pt. 18° C.).

In their chemical properties the ethylenes differ considerably from the paraffins. The former, on account of their unsaturated nature, form a number of addition products. Thus they combine directly with hydrogen to give paraffins, with halogens to give dihalogen derivatives of the paraffins, with halogen hydracids to give monohalogen paraffin derivatives, and with sulphuric acid to give alkyl-sulphuric acids, which are soluble in water.

$$C_2H_4 + H_2 \rightarrow C_2H_6 \qquad \text{Ethane.}$$
$$C_2H_4 + Cl_2 \rightarrow C_2H_4Cl_2 \qquad \text{Ethylene dichloride.}$$
$$C_2H_4 + HI \rightarrow C_2H_5I \qquad \text{Ethyl iodide.}$$
$$C_2H_4 + H_2SO_4 \rightarrow C_2H_5HSO_4 \qquad \text{Ethyl hydrogen sulphate.}$$
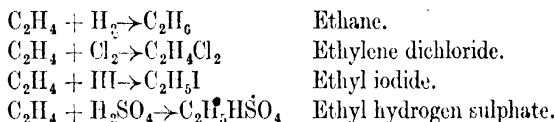
In the absence of water the olefines also unite directly with ozone, forming highly explosive ozonides of the general formula $C_nH_{2n}O_3$ which decompose in the presence of water.

The olefines further differ from the paraffins in their tendency to polymerise, especially in the presence of such substances as sulphuric acid or zinc chloride. Thus amylene, C_5H_{10}, gives rise to polymerides of the composition $C_{10}H_{20}$ and $C_{20}H_{40}$. Lastly, it should be noted that the olefines, unlike the paraffins, are readily oxidised by agents such as chromic acid or potassium permanganate.

3. DIOLEFINES

Although possessing the structure and characteristics of the olefines, the group of hydrocarbons known as the diolefines have the same general formula, C_nH_{2n-2},

[1] See Schütz, Buschmann and Wissebach, " Low Temperature Coal Tar and the Products of its Over-heating." *Ber.*, 1923, **56**, 1091.

as the acetylenes, to which reference is made below. Among the members of this group are :—

Allene (propadiene)	C_3H_4,	$CH_2 : C : CH_2$.
Divinyl (butadiene)	C_4H_6,	$CH_2 : CH \cdot CH : CH_2$.
Piperylene (pentadiene)	C_5H_8,	$CH_2 : CH \cdot CH : CH \cdot CH_3$
Diallyl	C_6H_{10},	$CH_2 : CH \cdot CH_2 \cdot CH_2 \cdot CH : CH_2$.

Inspection of the formulæ of these hydrocarbons shows that in each of them the characteristic ethylenic linkage $>C : C<$ occurs twice ; the substances consequently possess the properties of the ethylenes to an enhanced degree, and are not to be regarded as acetylenes. According to Jones and Wheeler[1] and Schutz,[2] diolefines result from the low temperature distillation of coal.

4. Acetylenes

As mentioned above, the homologous series of hydrocarbons known as the acetylenes have the general formula C_nH_{2n-2}. The series includes the following compounds :—

Acetylene C_2H_2,	$CH : CH \cdot$		B. Pt.	$- 82°$ C.
Allylene (methylacetylene) C_3H_4,	$CH : C \cdot CH_3$		,,	$- 23\cdot5°$ C.
Crotonylene C_4H_6,	$CH_3 \cdot C : C \cdot CH_3$		,,	$27°$ C.

Acetylene itself has been identified by Burgess and Wheeler[3] as a product of low temperature distillation.

The acetylenes being still more unsaturated than the olefines, are even more reactive than the latter. Thus, under suitable conditions they combine directly with two or four atoms of hydrogen or halogens, or with one or two molecules of halogen hydracids :—

$$C_2H_2 + H_2 \rightarrow C_2H_4 \qquad \text{Ethylene.}$$
$$C_2H_2 + 2H_2 \rightarrow C_2H_6 \qquad \text{Ethane.}$$
$$C_2H_2 + Br_2 \rightarrow C_2H_2Br_2 \qquad \text{Dibromethylene.}$$
$$C_2H_2 + 2Br_2 \rightarrow C_2H_2Br_4 \qquad \text{Ethylene tetrabromide.}$$
$$C_2H_2 + HBr \rightarrow C_2H_3Br \qquad \text{Vinyl bromide.}$$
$$C_2H_2 + 2HBr \rightarrow C_2H_4Br_2 \qquad \text{Ethylene dibromide.}$$
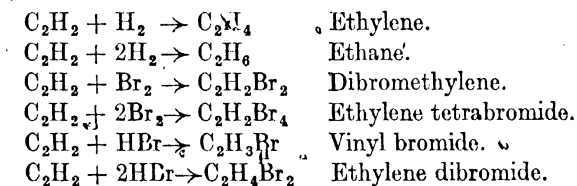
In the presence of dilute acids the acetylenes combine with the elements of water ; in this way acetylene yields acetaldehyde, and allylene gives acetone :—

$$CH : CH + H_2O \rightarrow CH_3 \cdot CHO.$$
$$CH_3 \cdot C : CH + H_2O \rightarrow CH_3 \cdot CO \cdot CH_3.$$
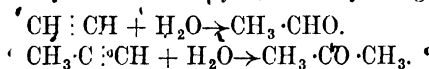
[1] Jones and Wheeler. "The Composition of Coal." *Trans. Chem. Soc.*, 1914, **105**, 140.
[2] Schutz. "Nature of the Hydrocarbons present in Low Temperature Tar Light Oil." *Brennstoff-chemie*, 1923, **4**, 84.
[3] Burgess and Wheeler. "The Volatile Constituents of Coal." *Trans. Chem. Soc.*, 1915, **107**, 1916, and subsequent papers.

The acetylenes also unite directly with ozone, giving ozonides readily decomposed by water. The tendency to polymerisation shown by the ethylenes is still more marked in the acetylenes. Thus, on heating, acetylene polymerises to benzene $(3C_2H_2 \rightarrow C_6H_6)$, and under appropriate conditions allylene becomes converted into mesitylene $(3C_3H_4 \rightarrow C_9H_{12})$.

Those of the acetylenes which contain the group $-C : CH$ possess the somewhat curious property of forming metallic derivatives such as copper acetylide, $Cu\cdot C : C\cdot Cu$, and silver acetylide, $Ag\cdot C : C\cdot Ag$; the acetylides of the heavy metals are dangerously explosive when dry.

5. NAPHTHENES OR PARAFFENES

The general formula for this group is, like that of the ethylenes, C_nH_{2n}; the molecules of these hydrocarbons are composed of varying numbers of $-CH_2-$ (methylene) groups, assembled in ring formation, and are hence known as polymethylenes. The following are members of this series :—

		B. Pt.
Trimethylene	$\begin{array}{c} CH_2 \\ \diagup \diagdown \\ CH_2 - CH_2 \end{array}$	$-35°$
Tetramethylene	$\begin{array}{c} CH_2 - CH_2 \\ \vert \qquad \vert \\ CH_2 - CH_2 \end{array}$	$12°$
Pentamethylene	$\begin{array}{c} CH_2 - CH_2 \\ \vert \qquad \diagdown CH_2 \\ CH_2 - CH_2 \end{array}$	$50°$
Hexamethylene	$\begin{array}{c} CH_2 - CH_2 \\ \diagup \qquad \diagdown \\ CH_2 \qquad CH_2 \\ \diagdown \qquad \diagup \\ CH_2 - CH_2 \end{array}$	$81°$
Heptamethylene	$(CH_2)_7$	$117°$

According to Jones and Wheeler[1] and Morgan and Soule,[2] members of the naphthene series are found in the products of low temperature distillation; while Pictet and Bouvier[3] claim to have identified naphthenes having the formulæ C_9H_{18}, $C_{10}H_{20}$, $C_{11}H_{22}$, $C_{12}H_{24}$, and $C_{13}H_{26}$. It should be noted that no substances are known,

[1] Jones and Wheeler "The Composition of Coal." *Trans. Chem. Soc.* 1914, **105**, 140.
[2] Morgan and Soule. "Examination of Low Temperature Coal Tars." *Journ. Ind. Eng. Chem.*, 1922, **15**, 693.
[3] Pictet and Bouvier. *Compt. rend.*, 1913, **157**, 1436; 1915, **160**, 629 and 926.

which contain more than nine carbon atoms united in ring formation; the last-mentioned compounds therefore doubtless have side chains, e.g. :—

$$C_{10}H_{20} = CH_2 \underset{CH_2---CH_2}{\overset{CH_2---CH_2}{\diagdown}} CH \cdot CH_2 \cdot CH_2 \cdot CH_2 \cdot CH_3.$$
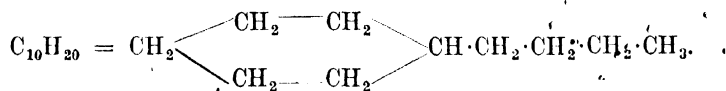
It is interesting to note that cyclohexane (hexamethylene) can be prepared by the direct union of hydrogen with benzene in the presence of metallic nickel (the Sabatier and Senderens reaction) :—

$$+ 3H_2 \rightarrow$$
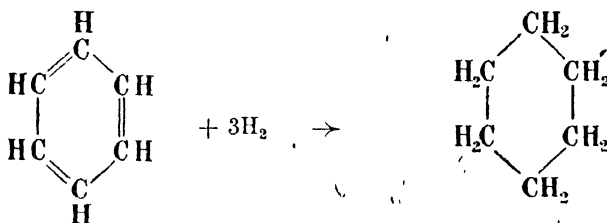
Many hydrocarbons derived from benzene—toluene, for instance, can be similarly reduced :—

$$+ 3H_2 \rightarrow$$
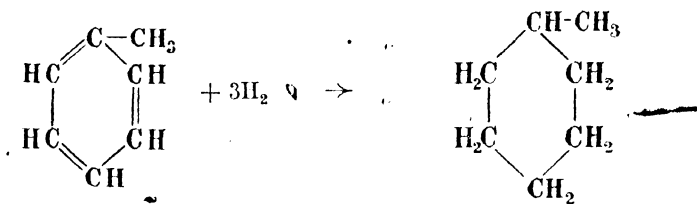
and in this way derivatives of cyclohexane are obtained.

Examination of the formulæ of the members of this series of hydrocarbons shows that they are saturated substances ; the carbon atoms in their molecules are singly linked to each other. In their properties they show a marked resemblance to the paraffins, and they may consequently be called cycloparaffins ; they differ very sharply from the straight chain olefines. Thus, unlike the latter, they do not form addition compounds with sulphuric acid and the halogens. The hydrocarbons derived from cyclohexane are the commonest and best known members of the cycloparaffin group.

6. Cyclopentadienes

The hydrocarbon cyclopentadiene, C_5H_6, may be regarded as derived from cyclopentane by the loss of four atoms of hydrogen, thus :—

$$\begin{matrix} CH_2 - CH_2 \\ CH_2 - CH_2 \end{matrix} \Big\rangle CH_2 \rightarrow \begin{matrix} CH = CH \\ CH = CH \end{matrix} \Big\rangle CH_2.$$
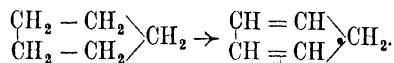
This compound has been identified in high temperature coal tar[1] and is said by Schütz, Buschmann and Wissebach[2] to be also a product of low temperature distillation. Other cyclodienes are probably also present as, indeed, the above authors suggest. Cyclopentadiene, which is a liquid boiling at 41° C., is a very reactive hydrocarbon whose general chemical nature is olefinic ; it readily combines, for instance, with bromine.

7. Benzene Hydrocarbons

The following table shows the hydrocarbons of the benzene series which are of importance from the low temperature distillation point of view :—

Benzene C_6H_6		.	.	M. Pt. + 5·4°	B. Pt.	80°
Toluene $C_6H_5 \cdot CH_3$	.	.	.	.	.	,,	111°
o-Xylene $CH_3 \cdot C_6H_4 \cdot CH_3$		.	.	.	.	,,	142°
m-Xylene	,,	,,	.	.	.	.	,,	139·9°
p-Xylene	,,	,,		.	.	.	,,	138°
Mesitylene	$C_6H_3(CH_3)_3$	.	.	.	.	,,	163°
Pseudocumene	,,	.	.	.	.	,,	169·8°
Durene $C_6H_2(CH_3)_4$		.	.	.	.	,,	190°

There is considerable controversy as to whether benzene, toluene and the three xylenes are formed during the low temperature distillation of coal ; it seems probable that they occur in very small quantity in isolated specimens.

Jones and Wheeler[3] identified only toluene, and that in traces.[4] Parr and Olin[5] state that they have identified all these five hydrocarbons. Parr and Olin[6] add that mesitylene is possibly also present ; while pseudocumene has been identified

[1] Ber., 1896, **29**, 552.
[2] Schütz, Buschmann and Wissebach. "Low Temperature Coal Tar and the Products of its Over-heating." Ber., 1923, **56**, 1091.
[3] Jones and Wheeler. "The Composition of Coal." Trans. Chem. Soc., 1914, **105**, 140.
[4] Cf. Pictet and Bouvier. "Sur la Distillation de la Houille sous Pression réduite." Compt. rend., 1913, **157**, 779 ; Schütz, Buschmann and Wissebach. "Low Temperature Coal Tar and the Products of its Over-heating." Ber., 1923, **53**, 1091.
[5] Parr and Olin. "The Coking of Coal at Low Temperature." Bull. **79**, Eng. Expt. Sta., Univ. of Illinois.
[6] Parr and Olin, loc. cit.

by Weissberger and Moehrle.[1] The authors last mentioned also identified[2] durene in a low temperature tar.

Benzene (C_6H_6) is the simplest aromatic hydrocarbon; in its molecule the carbon atoms form a six-membered ring, and each has attached to it a hydrogen atom. For a discussion of its structural formula the reader is referred to a textbook of organic chemistry. Representing its formula by the conventional hexagon, each angle of which portrays a carbon atom with one hydrogen atom attached to it, the *formulæ of the simplest members* of this group of hydrocarbons may be written as follows :—

CH₃.

Benzene   Toluene

CH₃   CH₃   CH₃

CH₃

CH₃

CH₃

o-Xylene   m-Xylene   p-Xylene

CH₃   CH₃   CH₃

CH₃   CH₃

H₃C   CH₃   H₃C

CH₃   CH₃   CH₃

Mesitylene   Pseudocumene   Durene
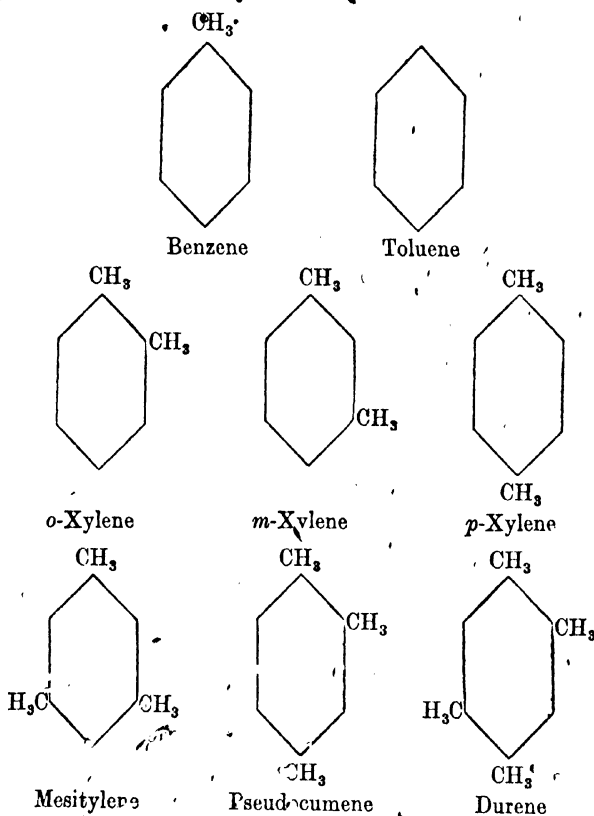
In these formulæ the $-CH_3$ groups each take the place of one hydrogen atom in the benzene nucleus.

[1] Weissberger and Moehrle. "Low Temperature Tar Oil.", *Brennstoffchemie*, 1923, 4, 81.
[2] Weissberger and Moehrle, *loc. cit.*

In chemical behaviour the benzene hydrocarbons are quite different from the other hydrocarbons we have already considered. The nuclear ring structure is very stable and persists unchanged in the presence of powerful reagents. These hydrocarbons are readily nitrated by strong nitric acid, and with concentrated or fuming sulphuric acid yield sulphonic acids—

$$C_6H_6 + HO \cdot NO_2 \rightarrow H_2O + C_6H_5 \cdot NO_2$$
$$\text{Nitrobenzene}$$

$$C_6H_6 + HO \cdot SO_2 \cdot OH \rightarrow H_2O + C_6H_5 \cdot SO_2OH$$
$$\text{Benzenesulphonic acid}$$
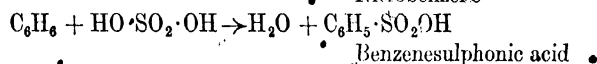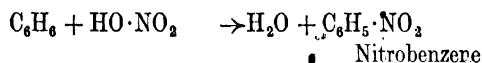
The hydrogen atoms of the benzene nucleus can be displaced, one by one, in this way, and mono-, di- and tri-derivatives are thus formed. Benzene is extremely difficult to oxidise; it is slowly attacked by potassium permanganate yielding formic and oxalic acids. The homologues of benzene are, however, more easily oxidised, the side-chain being converted into a carboxyl-group, e.g. :—

$$C_6H_5CH_3 + 3O \rightarrow C_6H_5COOH + H_2O$$
$$\text{Toluene} \qquad \text{Benzoic acid}$$
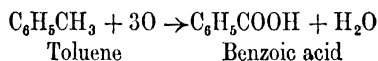
Benzene and its homologues can be partially or completely reduced; in the latter case hexahydrobenzene (cyclohexane) or its derivatives result.

Chlorine and bromine react with the benzene hydrocarbons in two ways. Benzene, for instance, when the reaction occurs in direct sunlight, yields the addition products $C_6H_6Cl_6$ and $C_6H_6Br_6$. In diffused light, however, and in the presence of a " halogen carrier " (iodine or antimony trichloride), substitution products, such as C_6H_5Cl, chlorobenzene, are obtained. Benzene and its homologues exhibit a number of other characteristic reactions which cannot be described here; sufficient has been said to indicate the marked difference there is between them and the hydrocarbons already discussed.

In the section dealing with the naphthenes it was pointed out that fully saturated six carbon ring compounds could be obtained by the complete reduction of the benzene nucleus. Besides these fully saturated compounds a number of intermediate reduction products may be formed; thus, from toluene, for example, dihydro- and tetrahydro-derivatives can be obtained, e.g. :—

 and
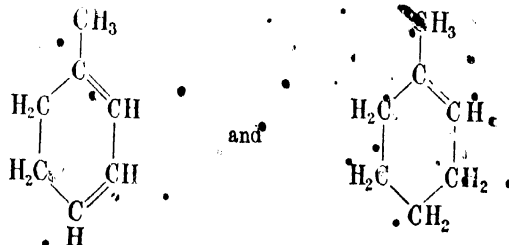
Substances of the first type, namely, dihydro-m-xylene (C_8H_{12}), dihydrotrimethyl-benzene (C_9H_{14}), dihydropseudocumene (C_9H_{14}) and dihydromesitylene (C_9H_{14}), have been found in low temperature tar by Pictet and Bouvier.[1] Again, Eckard[2] reports that substances $C_{10}H_{18}$ and $C_{11}H_{20}$ are present; these possibly contain a tetrahydrobenzene nucleus.

The chemical characteristics of the parent benzenes are considerably modified by the introduction of two or four hydrogen atoms; they no longer behave like the benzenes, nor are they fully saturated like the cyclohexanes previously mentioned.

8. NAPHTHALENE HYDROCARBONS

The parent hydrocarbon of this group, naphthalene $C_{10}H_8$, may be represented by the formula :—
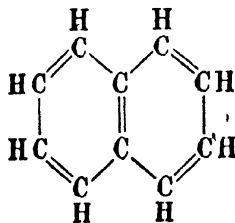
Naphthalene itself is a crystalline solid at ordinary temperatures (M. Pt. 79·6–79·8° C.; B. Pt. 218° C.); it is peculiar in its high volatility and its capacity for dissolving large quantities of air, when molten, which are given up on solidification. Several authors[3] agree that, although the homologues of naphthalene are present, naphthalene itself is absent from low temperature tars. Weissberger and Moehrle,[4] however, claim that it is present. Naphthalene is formed by strongly heating a number of substances, and this has been given as the cause of its presence in ordinary coal tar.

Three homologues of naphthalene have been discovered in low temperature tars; α-methylnaphthalene and β-methylnaphthalene were found by Fischer.[5] These substances may be represented by the following formulae :—

[1] Pictet and Bouvier. *Compt. rend.*, "Sur le Goudron du vide," 1913, **157**, 1436; "Sur les Hydrocarbures saturés du Goudron du vide," 1915, **160**, 629.

[2] Fromm and Eckard. "Lignite Low Temperature Tar." *Ber.*, 1923, **56**, 948.

[3] Jones and Wheeler. "The Composition of Coal." *Trans. Chem. Soc.*, 1914, **105**, 140; Parr and Olin. "The Coking of Coal at Low Temperature," *Bull.* 79, Eng. Expt. Sta., Univ. of Illinois; Fischer and Gluud. *Brennstoffchemie*, 1922, **3**, 57.

[4] Weissberger and Moehrle. "Low Temperature Tar Oils." *Brennstoffchemie*, **4**, 81.

[5] Fischer, Schrader and Zerbe. "On the absence of Naphthalene and the presence of Derivatives of Naphthalene in Tar obtained at a Low Temperature." *Ber.*, 1922, **55**, 57.

CH₃

α-Methylnaphthalene   β-Methylnaphthalene
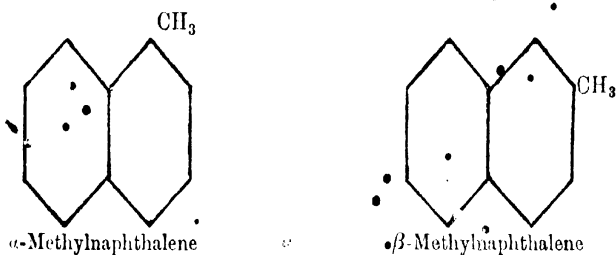
The first is an oil (Setting Pt. — 22° C. ; B. Pt. 241–242° C.) ; the second is a solid (M. Pt. 32° C. ; B. Pt. 241–242° C.). A third derivative of naphthalene, 1 : 6-dimethylnaphthalene, was reported by Weissberger and Moehrle.[1] It is an oil with a faint odour (B. Pt. 265° C.), and its formula may be written as :—
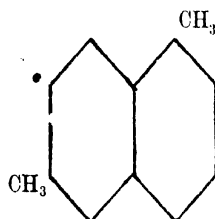
The chemical characteristics of the naphthalene group are very similar to those of the benzenes, but a larger number of derivatives is possible ; thus, for example, two monosubstituted methylnaphthalenes are cited above, whereas in the benzene series there is only one monomethyl-derivative, viz. toluene. The side-chains of the naphthalene homologues, like those of the benzene series, are easily oxidised to form acids.

Weissberger and Moehrle[2] have discovered a fully reduced naphthalene, deca-hydronaphthalene ($C_{10}H_{18}$), in low temperature tar. It is a liquid boiling at 187° C. Its chemical properties resemble those of the polymethylenes. This substance can be prepared synthetically from naphthalene by reduction by means of hydrogen in presence of nickel.[3]

[1] Weissberger and Moehrle. " Low Temperature Tar Oils." *Brennstoffchemie*, 1923, **4**, 81 ; also see Morgan and Soule. "Examination of Low Temperature Coal Tars." *Journ. Ind. Eng. Chem.*, 1923, **15**, 693.

[2] Weissberger and Moehrle. " Low Temperature Tar Oils." *Brennstoffchemie*, 1923, **4**, 81.

[3] Leroux. " Tétrahydrure et Décahydrure de Naphtaline." *Compt. rend.*, 1904, **139**, 672.

$$+ 5H_2 \rightarrow$$
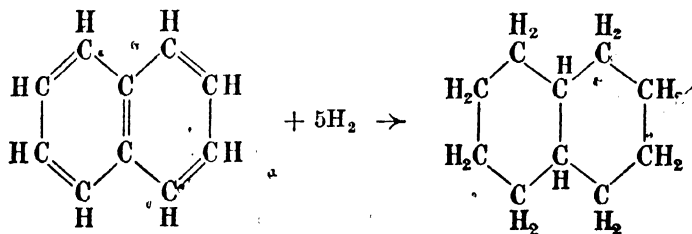
9. OTHER HYDROCARBONS

Anthracene ($C_{14}H_{10}$) was discovered in ordinary coal tar by Fritzsche in 1857, but was definitely stated to be absent from low temperature tar by Parr and Olin.[1] It may be represented by the formula :—

or
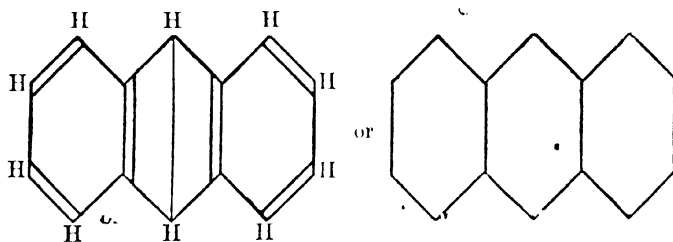
It is a solid (M. Pt. 216° C.; B. Pt. 351° C.), and is very similar to naphthalene in its chemical properties. Like the latter substance, it is formed pyrogenetically from a large number of substances, such as toluene, benzene, turpentine, oil, etc. ,

Acenaphthene ($C_{12}H_{10}$) occurs in ordinary coal tar in appreciable quantities, and was first found there by Berthelot in 1867.[2] It does not appear to have been detected in low temperature tar, but the fully reduced substance, perhydroacenaphthene, has been found by Weissberger and Moehrle,[3] the boiling-point of the latter substance being 235° C. The formation of perhydroacenaphthalene from acenaphthalene may be represented by the equation :—

[1] Parr and Olin. "The Coking of Coal at Low Temperature." *Bulletin* 97, Eng. Exp. Sta., Univ. of Illinois.

[2] Berthelot, *E. J. Chemie*, 1867, 714.

[3] Weissberger and Moehrle. "Low Temperature Tar Oils." *Brennstoffchemie*, 1923, **4**, 81.

$$CH_2\text{---}CH_2 \quad + 5H_2 \quad \rightarrow$$
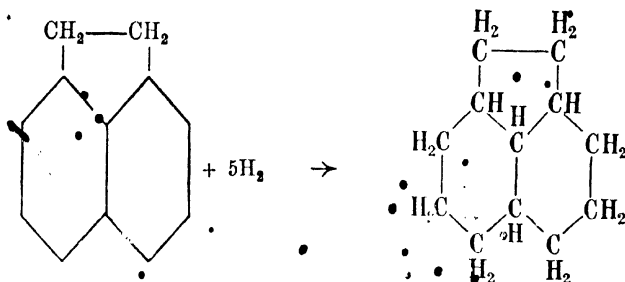
Dodecahydrodiphenyl has also been found by Weissberger and Moehrle. It is a liquid boiling at 235° C. Synthetically it can be prepared by the reduction of diphenyl by the Sabatier and Senderens reaction :—

$$+ 6H_2 \quad \rightarrow$$
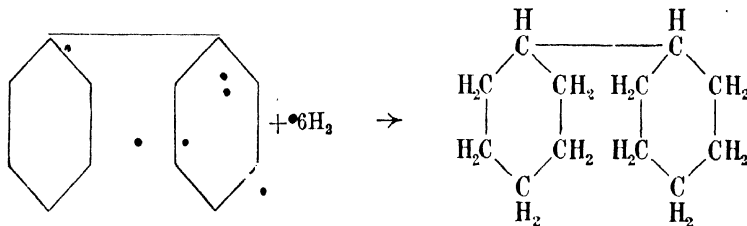
Fluorene, $C_{13}H_{10}$ or , has been discovered in ordinary coal tar,

and is separated by means of its potassium salt (A.G. für Teer- und Erdöl-Industrie, D.R.P. 124,150). It has not been found in low temperature tar. The parent substance is a colourless crystalline solid (M. Pt. 115°; B.·Pt. 295° C.). Perhydrofluorene, the fully reduced compound, has been found in low temperature tar by Weissberger and Moehrle ; it is a liquid boiling at 25b 258° C.

Styrolene (C_8H_8) has not been stated to be present in low temperature tar, but is found in ordinary coal tar. Like other unsaturated substances styrolene forms additive compounds with the halogens, etc. :—

$$\text{---CH : CH}_2 \quad + Br_2 \quad \rightarrow \quad \text{CH Br.CH}_2Br.$$
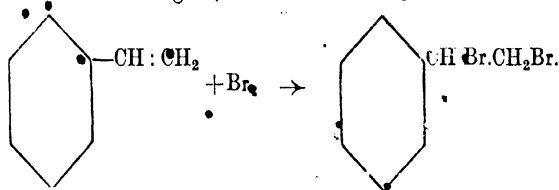
It is a liquid (B. Pt. 145-146° C.) which polymerises on standing to form meta-styrolene.

Indene (C_9H_8) itself has not been found in low temperature tar, but the presence of homologues is claimed by Weissberger and Moehrle.[1] The structure of this hydrocarbon may be represented as :—

$$C_6H_4 \diamondsuit \begin{matrix} CH \\ CH \\ CH_2 \end{matrix}$$
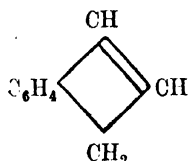
It is a liquid (Setting Pt. − 2° C. ; B. Pt. 181·3° C.), and was discovered in ordinary coal tar by Kramer and Spilker.[2] It polymerises easily and exhibits the characteristic properties of an unsaturated compound.

B. COMPOUNDS CONTAINING OXYGEN

Water is always present in the products formed during the distillation of coals which contain varying amounts of water held mechanically ; this, however, does not constitute the only source of the water produced ; it is also formed by the chemical reactions taking place in the retort.

The chief oxygen-containing compounds with which we have to deal are the alcohols, aldehydes, ketones and acids.

ALCOHOLS

Three possible types of alcohols must be considered :—

(a) Primary alcohols, which have a hydroxyl-group directly attached to a carbon atom which itself is attached to two hydrogen atoms, i.e. $R-C{<}^H_H OH$. This type of alcohol is unimportant from the present point of view. On oxidation it yields either an aldehyde $\left(R-C{<}^H_O\right)$ or an acid $\left(R-C{<}^{OH}_O\right)$, according to the extent to which oxidation proceeds.

[1] Weissberger and Moehrle, Brennstoffchemie, 1923,4, 81.
[2] Kramer and Spilker, Ber., 1890, 23, 3276 ; 1909, 42, 573.

(b) Secondary alcohols, in which the hydroxyl-group is attached to a carbon atom possessing only one hydrogen atom, i.e. $\begin{smallmatrix} R \\ R \end{smallmatrix}>C<\begin{smallmatrix} H \\ OH \end{smallmatrix}$. Few examples of secondary alcohols appear to have been isolated from low temperature tar ; possible exceptions are the menthol-like compounds noted by Pictet and Bouvier, and the hydrogenated cresol by Pictet, Kaiser and Labouchère. On oxidation, secondary alcohols yield ketones :—

$$2\begin{smallmatrix} R \\ R \end{smallmatrix}>C<\begin{smallmatrix} H \\ OH \end{smallmatrix} + O_2 \rightarrow 2\begin{smallmatrix} R \\ R \end{smallmatrix}>C \cdot O + 2H_2O$$
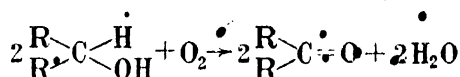
(c) Tertiary alcohols, in which the hydroxyl-group is attached to a carbon atom having no hydrogen atom. Oxidation of this class of alcohol cannot be brought about without rupture of the molecule. This is the most important type of alcohol in coal tar chemistry, if we regard phenol and the cresols as belonging to it.

It should be noted that the hydroxyl-group, when directly attached to an aromatic nucleus, such as benzene or naphthalene, displays acidic properties ; thus a sodium salt is formed by interaction of phenol with sodium hydroxide. This property is made use of in the separation of the carbolic oils ; they are obtained by the extraction of a fraction (more or less restricted in range of distillation) by means of sodium hydroxide solution. The sodium compounds, being soluble in water, pass into the aqueous layer and can be recovered by the addition of acid. The separation of phenol from cresol is based on the stronger acidity of the former.

The following phenolic substances have been found in low temperature tar :—

Substance.				M. Pt.	B. Pt.
Phenol	.	.	.	43° C.	181° C.
o-Cresol	.	.	.	30° C.	188° C.
m-Cresol	.	.	.	4° C.	200° C.
p-Cresol	.	.	.	36° C.	202° C.
1 : 2 : 4-Xylenol	.	.	.	65° C.	225° C.
Trimethylphenols	.	.	.	—	—
Tetramethylphenols	.	.	.	—	—
Pentamethylphenol	.	.	.	—	—
Catechol	.	.	.	104° C.	245° C.

Chemically these phenolic substances are very similar ; throughout the series the hydroxyl-group exhibits acidity, this property being modified in degree by the influence of the methyl groups. The above substances may be represented by the formulæ :—

CH_3 CH_3 CH_3 CH_3

OH OH OH CH_3

OH OH

Phenol o-Cresol m-Cresol p-Cresol 1 : 2 : 4-Xylenol

OH OH

CH_3 CH_3 OH

CH_3 CH_3

CH_3

Pentamethylphenol Catechol
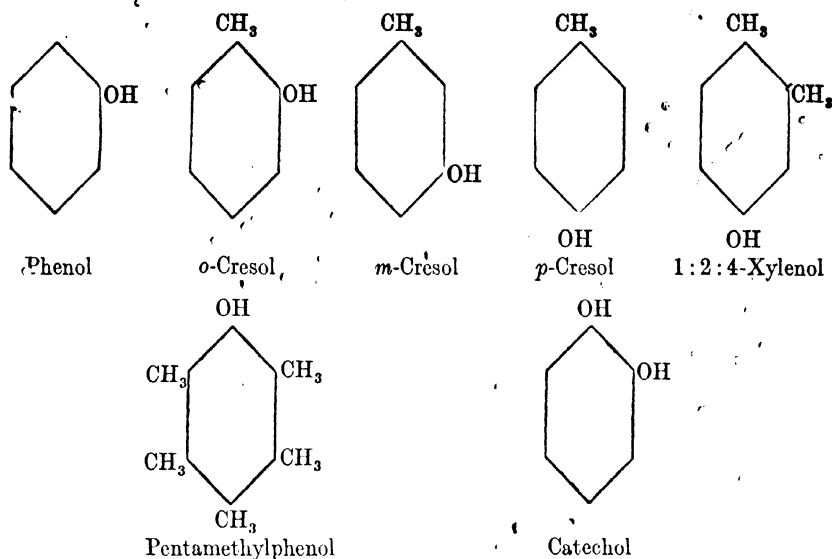
Phenol (C_6H_5OH) occurs in ordinary coal tar, but its presence in low temperature tar is much disputed. Pictet and Bouvier[1] support the view that it is not a product of low temperature distillation, but they state that they found it in similar tars which had stood for five years. Fischer[2] states that phenol is not always present ; but Morgan and Soule[3] claim to have found it in tar from carbocoal.

The latter authors found the cresols in the same material, and Jones and Wheeler[4] claim that cresols are present in low temperature tars ; Pictet and Bouvier[1] detected them in tar that had stood for five years. Gluud and Breuer[5] have also found cresols.

Xylenols ($C_8H_{10}O$) have been found by various authors.[6] Pictet and Bouvier[6] isolated 1 : 2 : 4-xylenol from tar that had stood for five years.

Trimethylphenols ($C_9H_{12}O$) have been identified in low temperature tar by Gluud and Breuer,[5] Pictet, Kaiser and Labouchère,[6] and Eckard.[6]

The presence of tetramethyl and pentamethyl phenols is also claimed by Pictet, Kaiser and Labouchère.[6]

[1] Pictet and Bouvier. Compt. rend., 1913, 157, 779.
[2] Fischer. Ges. Abh. zur Kenntnis der Kohle, 1918, 3, 89 ; Brennst. ffchemie, 1920, 1, 31, 47.
[3] Morgan and Soule. "Studies in the Carbonisation of Coal." Chem. Met. Eng., 1922, 26, 926.
[4] Jones and Wheeler. "The Composition of Coal." Trans. Chem. Soc., 1914, 105, 140.
[5] Gluud and Breuer. Ges. Abh. zur Kenntnis der Kohle, 1918, 2, 236.
[6] Morgan and Soule, loc. cit. ; Fromm and Eckard. "Lignite Low Temperature Tar." Ber., 1923, 56, 948 ; Gluud and Breuer, loc. cit. ; Jones and Wheeler, loc. cit. ; Pictet, Kaiser and Labouchère. Compt. rend., 1917, 165, 113 ; Pictet and Bouvier, loc.cit.

Gluud and Breuer[1] have reported that catechol (o-dihydroxy-benzene) is present in low temperature tar.

Besides the true phenolic substances a hydrogenated cresol, hexahydro-p-cresol has been found by Pictet, Kaiser and Labouchère.[2] By inspection of its structural formula—

$$
\begin{array}{c}
CH_3 \\
| \\
CH \\
H_2C \quad CH_2 \\
| \qquad | \\
H_2C \quad CH_2 \\
C \\
HOH
\end{array}
$$
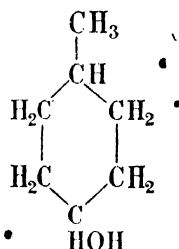
it will be seen that this substance is a secondary alcohol of the cyclohexane series. As such its nature is more that of an aliphatic alcohol than of a phenol. The substance is a liquid boiling at 174° C.

ALDEHYDES

An aldehyde may be represented by the general formula $R-C\diagdown^{H}_{O}$ where R may be a simple chain radicle or a radicle of the benzene type. In the former case the aldehydic group displays great chemical activity. Thus, for instance, acetaldehyde $\left(CH_3-C\diagdown^{H}_{O}\right)$ is readily oxidised in the air to form acetic acid $\left(CH_3-C\diagdown^{O}_{OH}\right)$; it causes, by reduction, the separation of metallic silver from ammoniacal solution of silver salts, and it further forms a number of additive compounds.

In the presence of small quantities of acids or of certain salts, acetaldehyde is converted into a polymeride, paraldehyde $(C_2H_4O)_3$. Acetaldehyde itself has not been found in low temperature tar; but paraldehyde has been claimed to be present by Schutz.[3]

Paraldehyde is a colourless liquid at ordinary temperatures (M. Pt. 12·6° C.; B. Pt. 124° C.). It does not exhibit the reactivity of acetaldehyde. When distilled with sulphuric acid, acetaldehyde is generated.

[1] Gluud and Breuer. Ges. Abh. zur Kenntnis der Kohle, 1918, 2, 236.
[2] Morgan and Soule, loc. cit.; Fromm and Eckard. "Lignite Low Temperature Tar." Ber. 1923, 56, 948; Gluud and Breuer, loc. cit.; Jones and Wheeler, loc. cit.; Pictet, Kaiser and Labouchère. Les Alcools et les Bases du Goudron du ville. Compt. rend., 1917, 165, 113; Pictet and Bouvier, loc. cit.
[3] Schutz. Brennstoffchemie, 1923, 4, 84.

KETONES

The general formula of ketones may be written as $\overset{R}{\underset{R}{>}}C = O$. It will be seen that they bear a close relationship to aldehydes, the hydrogen atom in the latter being replaced in ketones by a radicle such as $- CH_3$, $- C_2H_5$ or $- C_6H_5$. To some extent the properties of the ketones resemble those of the aldehydes ; but the former are not so easily oxidised, and consequently, in some directions, they are less reactive ; thus they are not capable of reducing alkaline silver salt solutions.

Acetone ($CH_3 \cdot CO \cdot CH_3$) is the simplest ketone, and was obtained from ordinary coal tar by Housler[1] ; while methylethylketone ($CH_3 \cdot CO \cdot C_2H_5$) was found in the same material by Schulze.[2] Both substances have been found in low temperature tar, the first by Broche,[3] and the second by Schütz, Buschmann and Wissebach.[4]

ACIDS

Although acetic acid ($CH_3 \cdot COOH$) and benzoic acid ($C_6H_5 \cdot COOH$) have been identified in ordinary coal tar, no reference to these or other organic acids is made in the literature of low temperature carbonisation.

C. COMPOUNDS CONTAINING SULPHUR

So far as the literature shows, the sulphur-containing compounds in the low temperature distillation products form no definite group or series ; brief reference is made below to the few individual substances which have been noted.

Hydrogen Sulphide (H_2S). Burgess and Wheeler[5] note the occurrence of this gas.

Carbon Disulphide (CS_2). Jones and Wheeler[6] state that this does not occur in low temperature gas ; but Schütz, Buschmann and Wissebach[7] claim to have found traces.

Methyl Mercaptan ($CH_3 \cdot SH$) and Dimethyl Sulphide ($(CH_3)_2S$). Both these substances have been noted by Schütz, Buschmann and Wissebach[8] as products of low temperature distillation. The former, the first member of the group of thioalcohols (e.g. alcohols in which sulphur takes the place of oxygen), is an evil-

[1] Housler.   *Ber.*, 1895, **28**, 488.
[2] Schulze.   *Ber.*, 1887, **20**, 411.
[3] Low Temperature Tar obtained from the Zeche Fürst Hardenberg Coal, and in particular, the content of Benzene, Phenol and Acetone.   *B. r.*, 1923, **56**, 1787–91.
[4] Schütz, Buschmann and Wissebach.   "Low Temperature Coal Tar and the Products of its Over-heating."   *Ber.*, 1923, **56**, 1091.
[5] Burgess and Wheeler.   "The Volatile Constituents of Coal."   *Trans. Chem. Soc.*, 1915, **107**, 1916, and subsequent papers.
[6] Jones and Wheeler.   "Composition of Coal."   *Trans. Chem. Soc.*, 1914, **105**, 140.
[7] Schütz, Buschmann and Wissebach.   "Low Temperature Coal Tar and the Products of its Over-heating."   *Ber.*, 1923, **56**, 1091.
[8] Schütz, Buschmann and Wissebach, *loc. cit.*

smelling liquid, lighter than water, which boils at the low temperature of $+ 6°$ C. The mercaptans are characterised by the formation of metallic derivatives, in which the metal takes the place of hydrogen. Thus ethyl mercaptan, or mercap. , as it is commonly called ($C_2H_5 \cdot SH$) gives sodium mercaptide ($C_2H_5 \cdot S Na$) and mercury mercaptide ($C_2H_5S)_2Hg$. A tolyl mercaptan ($CH_3 \cdot C_6H_4 \cdot SH$) has been identified in low temperature tar by Schutz.[1]

In the thioethers, of which dimethyl sulphide is the first member, sulphur takes the place of oxygen in common ethers, e.g. ethyl ether $(C_2H_5)_2O$, diethyl sulphide $(C_2H_5)_2S$.

The thioethers form derivatives with certain metallic salts by direct addition (e.g. $(C_2H_5)_2S \cdot HgCl_2$), are somewhat readily oxidised and unite directly with the halogens ; the tendency of the sulphur atom to pass from the di- to the tetra-valent state is the keynote to their chemical behaviour. Dimethyl-sulphide is a liquid (B. Pt. $37°$ C.) having an unpleasant and, at the same time, ether-like smell.

Thiophene. C_4H_4S occurs in high temperature tars ; its presence in low temperature tars is, however, problematical.[2] The substance is a colourless liquid ,with a faint smell resembling that of benzene, and boils at nearly the same temperature ($84°$ C.) as the latter. Its structural formula is—
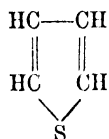
and, since the ring-structure is not composed exclusively of carbon atoms, it is known as a *heterocyclic* compound. When a minute quantity of thiophene is mixed with a few ccs. of strong sulphuric acid containing a crystal of isatin in solution, a deep blue colouration is developed ; the colour is due to the formation of indophenine, and the reaction is utilised as a delicate test for thiophene. In chemical behaviour thiophene does not in the least resemble the thioethers (*vide supra*) ; towards reagents such as sulphuric and nitric acids it behaves in a similar way to benzene and its derivatives.

D. COMPOUNDS CONTAINING CHLORINE

These can be dismissed by the brief statement that hydrochloric acid (HCl) and ammonium chloride (NH_4Cl) have been identified as products of low temperature distillation by Jones and Wheeler.[3]

[1] Schütz. "Low Temperature Tar." *Ber.*, 1923, **56**, 162.
[2] See Morgan and Soule. *Chem. and Met. Eng.*, 1922, **26**, 977.
[3] Jones and Wheeler. "The Composition of Coal." *Trans. Chem. Soc.*, 1914, **105**, 140.

E. COMPOUNDS CONTAINING NITROGEN

So far as low temperature tars are concerned, the nitrogenous substances which have so far been identified are, with one exception, basic in character. The exception is methyl cyanide (CH_3CN), which was identified by Schütz, Buschmann and Wissebach.[1] Methyl cyanide, or acetonitrile, as it is more commonly called, is a colourless combustible liquid (B. Pt. 82° C.). On heating with acids or alkalis it is hydrolysed, yielding ultimately acetic acid.

$$CH_3 \cdot CN + 2H_2O \rightarrow CH_3 \cdot COOH + NH_3.$$
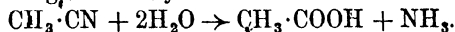
Under appropriate conditions it is reduced by hydrogen to ethylamine :—

$$CH_3CN + 2H_2 \rightarrow CH_3 \cdot CH_2 \cdot NH_2.$$

Of the basic nitrogenous substances ammonia is the simplest; many authors, e.g. Burgess and Wheeler,[2] refer to its occurrence. Its nature and properties are so well known that a description of them here is unnecessary. The principal nitrogenous bases in low temperature tars to which reference is made in the literature are shown below :—
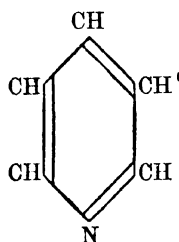
Pyridine

			B. Pt.
Pyridine C_5H_5N	.	. (M. Pt–42°)	115·5°
Picoline $C_5H_4N(CH_3)$	.	. α-isomer	129°
,, ,,	.	. β-isomer	144–147°
,, ,,	.	. γ-isomer	142·5–144·5°
Ethylpyridine $C_5H_4N(C_2H_5)$	.	. α-isomer	148·5°
Collidine (1 : 3 : 5 trimethylpyridine) $C_5H_2N(CH_3)_3$			172°
Hydroquinoline.			
Methylhydroquinoline.			
Ethylhydroquinoline.			
Isohydroquinoline.			
Toluidines $CH_3 \cdot C_6H_4 \cdot NH_2$	.	. o-isomer	199·7°
,, ,,	.	. m-isomer	203·3°
,, ,,	.	. p-isomer (M. Pt. 45°)	200·2°

[1] Schütz, Buschmann and Wissebach. "Low Temperature Coal Tar and the Products of its Over-heating." *Ber.*, 1923, **56**, 1051.

[2] Burgess and Wheeler. "The Volatile Constituents of Coal." *Trans. Chem. Soc.*, 1915, **107**, 1916, and later papers.

Pyridine. Jones and Wheeler[1] found pyridine bases in low temperature tars in traces only ; while Morgan and Soule[2] (see also Eckard[3]) identified pyridine in the tar from carbocoal. Pyridine is a colourless liquid with a peculiar smell ; it is freely miscible with water, and the solution is strongly alkaline. The substance is remarkable for its indifference to chemical reagents. It is a tertiary base and forms salts with acids (e.g. pyridine hydrochloride, C_5H_5N,HCl), which are usually soluble in water.

Picolines. The three isomeric methylpyridines are known as picolines ; their formulæ are as follows :—

α-picoline   β-picoline   γ-picoline
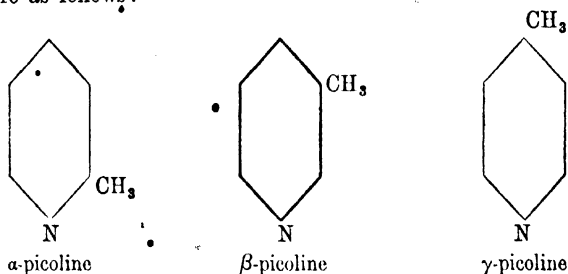
The picolines are liquids very similar in character to pyridine ; their occurrence has been noted by Morgan and Soule[2] and Eckard.[3] The three isomeric ethylpyridines are also known, and again are similar in behaviour to pyridine ; the occurrence of these substances is recorded by Morgan and Soule.[2]

1·3·5 Collidine. For the occurrence of this substance, see Gollmer.[4] Its formula is :—
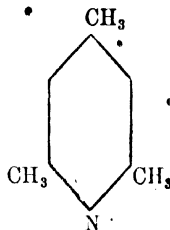
Pictet, Kaiser and Labouchère[5] note the presence of a hydroquinoline and of an isohydroquinoline ; while methyl- and ethyl-hydroquinoline have been identified in low temperature tars by Morgan and Soule.[2]

[1] Jones and Wheeler. " The Composition of Coal." *Trans. Chem. Soc.*, 1914, **105**, 140.
[2] Morgan and Soule. *Chem. and Met. Eng.*, 1922, **26**, 975.
[3] Fromm and Eckard. " Lignite Low Temperature Tar." *Ber.*, 1923, **56**, 948.
[4] Gollmer. " Basic Compounds in Low Temperature Coal Tar." *Brennstoffchemie*, 1923, **4**, 19.
[5] Pictet, Kaiser and Labouchère. *Compt. rend.*, 197, 105, 113.

Toluidines. The formulæ of the three isomeric toluidines, which are methyl-anilines, are as follows :—
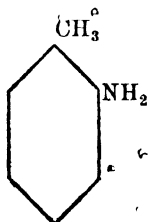
o-toluidine	m-toluidine	p-toluidine
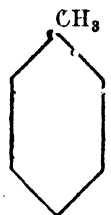
Pictet, Kaiser and Labouchère[1] record the presence of primary bases, apparently toluidines, in low temperature tars. The toluidines are the homologues of aniline ($C_6H_5NH_2$), which they greatly resemble in all their chemical properties. They are basic, neutralising acids and forming salts which are frequently soluble in water (e.g. toluidine hydrochloride, $CH_3 \cdot C_6H_4 \cdot NH_2, HCl$). Their reactions are numerous and important.

To complete the account of the nitrogenous basic substances it should be added that Pictet and Bouvier[2] have stated that secondary bases (e.g. of the type $\frac{R}{R_1}{>}NH$) were present in tars which they examined.

[1] Pictet, Kaiser and Labouchère. *Compt. rend.*, 1917, **165**, 113.
[2] Pictet and Bouvier. *Compt. rend.*, 1913, **157**, 779; also J. J. Morgan and R. P. Soule. "Examination of Low Temperature Coal Tars." *Ind. Eng. Chem.*, 1923, **15**, 587–91.

CHAPTER VII

THE EFFECT OF HEAT UPON COAL

INTRODUCTORY

MUCH experimental work has been undertaken to determine the effects of heat upon coal, not only with a view to obtaining or deducing information upon the constituents in coal, but also as a guide to the commercial carbonisation of coal at various temperatures. It is desired in this chapter to consider the effect of heat on coal from the latter standpoint.

The three principal products obtained by low temperature processes of carbonisation are coke, oils and gas, and the conditions must be fixed so that the quality and quantity of the yield are optimum, with particular reference to their total commercial value. It has already been indicated that, by increasing the temperature of carbonisation, the hardness of the coke and the yield of gas are in general improved, but that the quality of the gaseous products and the quantity of the oil are decreased. For low temperature processes a compromise must therefore be effected in fixing the temperature of carbonisation. This must be high enough to leave only sufficient residue of volatile matter in the coke to ensure its ease in kindling and free-burning properties and, at the same time, not so low as to invalidate the strength of the solid residue. The temperature must not be so high as to reduce the rich quality of the gas or the optimum of the oil yield. The particular problems to be discussed in this chapter are therefore :—

    A.   The most suitable temperature of carbonisation for low temperature processes.

    B.   The time required for carbonisation.

    C.   The amount of heat required for carbonisation.

A. THE TEMPERATURE OF CARBONISATION FOR LOW TEMPERATURE PROCESSES

SOME RESULTS OF EXPERIMENTAL RESEARCH

Coal is a very complex substance, and the products of decomposition vary considerably when it is heated to different temperatures. As the composition of coal also varies over a wide range, it is impossible to make sound generalisations of the yield at any temperature which would be applicable to all types of coal. The following results of experimental investigations in the laboratory on carbonaceous materials at various temperatures, but more especially the lower temperatures, will, however, first be considered.

Börnstein in 1906[1] published the results of researches on the low temperature

---

[1] Börnstein, E. " Decomposition of Solid Fuels at gradually increasing temperatures," *Journ. f. Gasbeleuchtung*, **49**, 627–30, 648–52, 667–71.

distillation of wood, peat and lignite. The ultimate analysis of each material is given in Table XIV, and the composition of gas evolved at various temperatures is given in Table XV.

TABLE XIV

ULTIMATE ANALYSIS OF WOOD, PEAT, AND LIGNITE
IN BÖRNSTEIN'S EXPERIMENTS

	Wood.	Peat.	Lignite.
Carbon	48·75	43·76	63·44
Hydrogen	6·48	4·16	5·05
Oxygen	40·75	24·57	21·70
Nitrogen	0·63	2·30	0·83
Sulphur	0·08	0·24	0·45
Ash	0·10	7·94	6·22
Moisture	3·21	17·03	1·81

TABLE XV

GASES EVOLVED FROM WOOD PEAT AND LIGNITE (BÖRNSTEIN)

	250°C.—300°C.	300° C.—350° C.			350° C.—400° C.			400° C.—450° C.		
	Lignite.	Wood.	Peat.	Lignite.	Wood.	Peat.	Lignite.	Wood.	Peat.	Lignite.
Carbon Dioxide	91·4	53·5	89·2	90·9	55·0	63·8	69·6	28·0	55·4	27·8
Carbon Monoxide	6·4	27·7	10·1	7·6	32·6	7·2	15·8	29·0	12·3	15·1
Methane	1·1	14·9	—	—	7·9	25·5	3·2	20·6	25·4	21·2
Olefines	0·7	0·2	0·3	1·0	1·5	0·4	2·5	5·0	3·7	3·9
Hydrogen	0·3	3·7	0·3	0·5	3·0	3·1	8·8	17·3	3·1	11·9
Sulphur compounds	—	—	—	0·7	—	—	0·5	—	—	0·4

Börnstein also carbonised eight samples of coal at a temperature of 450° C. The proportion of the products varied considerably, but the following may be quoted :—

ULTIMATE ANALYSIS OF MOISTURE-FREE AND ASH-FREE COAL

C.	H.	O.	N.	S.
85·09	4·82	7·95	1·08	1·06

PERCENTAGE COMPOSITION OF GASES

Carbon Dioxide.	Carbon Monoxide.	Methane.	Ethane.	Olefines.	Hydrogen.
3·8	3·8	54·5	15·2	5·6	17·3

Unfortunately the quantity of the gases distilled from the substances investigated by Börnstein is not given.

A further contribution to this subject has also been made by Porter and Ovitz.[1] Ten grammes of non-coking coal (Illinois) and of coking coal (Pennsylvania) were progressively heated in an atmosphere of nitrogen to temperatures varying from 390° C. to 1026° C., and the gases given off were measured and analysed. The ultimate analysis of the coals on a moisture-free basis are given in Table XVI, and particulars of the gases evolved are given in Tables XVII and XVIII. The volumes of the gases evolved from the ten grammes of coal at the various temperatures are given in Tables XIX and XX and are shown in Figs. 4 and 5.

TABLE XVI

ULTIMATE ANALYSIS OF ILLINOIS AND PENNSYLVANIA COALS
(PORTER AND OVITZ)

	Illinois.	Pennsylvania.
Carbon	67·87	78·00
Hydrogen	5·44	5·24
Oxygen	19·47	7·47
Nitrogen	1·34	1·23
Sulphur	0·46	0·95
Ash	5·42	7·11

TABLE XVII

GASES EVOLVED FROM ILLINOIS COAL
(PORTER AND OVITZ)

Temperature of furnace °C.	500°	600°	700°	800°	900°	1000°	1100°
Highest temperature reached in coal °C.	390°	480°	585°	685°	811°	920°	1026°
Volume of gas evolved c.c. (from 10 grammes of coal)	197	535	980	1550	2335	2700	3120
Analysis (calculated nitrogen free) :—							
CO_2	23·8	7·6	6·4	3·9	2·5	2·7	1·8
Illuminants	6·5	5·0	4·1	3·3	2·2	3·7	4·0
CO	16·5	16·1	21·1	10·9	15·2	15·1	16·1
CH_4, C_2H_6, etc.	49·5	55·0	41·5	34·4	27·8	23·1	19·4
H_2	3·7	16·3	26·9	41·5	51·3	55·4	58·7

[1] Porter and Ovitz. "The Volatile Matter of Coal." Bulletin I, U.S.A. Bureau of Mines, 1910, 1–56.

TABLE XVIII

GASES EVOLVED FROM PENNSYLVANIA COAL (PORTER AND OVITZ)

Temperature of furnace °C.	500°	600°	700°	800°	900°	1000°	1100°
Highest temperature reached in coal °C.	390°	474°	589°	705°	812°	922°	1010°
Volume of gas evolved c.c. (from 10 grammes of coal)	161	718	1220	1723	2080	2900	3530
Analysis (calculated nitrogen free) :—							
CO_2	15·9	4·2	3·2	2·0	1·1	1·2	1·0
Illuminants	9·1	7·1	4·3	4·5	4·8	4·6	5·2
CO	7·8	6·0	6·3	7·2	7·4	6·4	7·3
CH_4, C_2H_6, etc.	63·3	64·4	55·8	47·0	33·2	29·0	26·3
H_2	3·9	18·3	30·4	39·3	53·5	58·8	60·2

TABLE XIX

VOLUMES OF GASES EVOLVED FROM ILLINOIS COAL IN C.C. PER TEN GRAMMES

Temperature °C.	390°	480°	585°	686°	811°	930°	1026°
CO_2	47·0	40·7	62·7	60·5	58·4	72·9	56·2
Illuminants	12·7	26·8	40·2	51·1	74·7	99·9	124·8
CO	32·5	86·1	206·8	262·0	354·9	407·7	502·3
CH_4, C_2H_6, etc.	97·5	294·2	406·7	533·2	649·1	623·7	605·3
H_2	7·3	87·2	263·6	643·2	1197·9	1495·8	1821·4
	197·0	535·0	980·0	1550·0	2335·0	2700·0	3120·0

TABLE XX

VOLUMES OF GASES EVOLVED FROM PENNSYLVANIA COAL IN C.C. PER TEN GRAMMES

Temperature °C.	390°	474°	589°	705°	812°	922°	1010°
CO_2	25·6	30·1	39·0	34·5	22·9	34·8	35·3
Illuminants	14·6	51·0	52·5	77·5	99·8	133·4	183·6
CO	12·6	43·0	76·9	124·1	133·9	185·6	257·7
CH_4, C_2H_6, etc.	102·0	462·4	680·7	809·8	690·6	841·0	928·4
H_2	6·2	131·5	370·9	677·1	1112·8	1705·2	2125·0
	161·0	718·0	1220·0	1723·0	2080·0	2900·0	3530·0

An examination of the products of carbonisation at 600° C. from typical coals in the low temperature assay apparatus described on page 60 has been made by Gray and King. In this method a 15-gramme sample contained in a glass tube was inserted to the full extent in the furnace at a temperature of 300° C. The temperature was then slowly raised to 600° C. during one hour, after which it was maintained at this level for a further period of one hour. As a
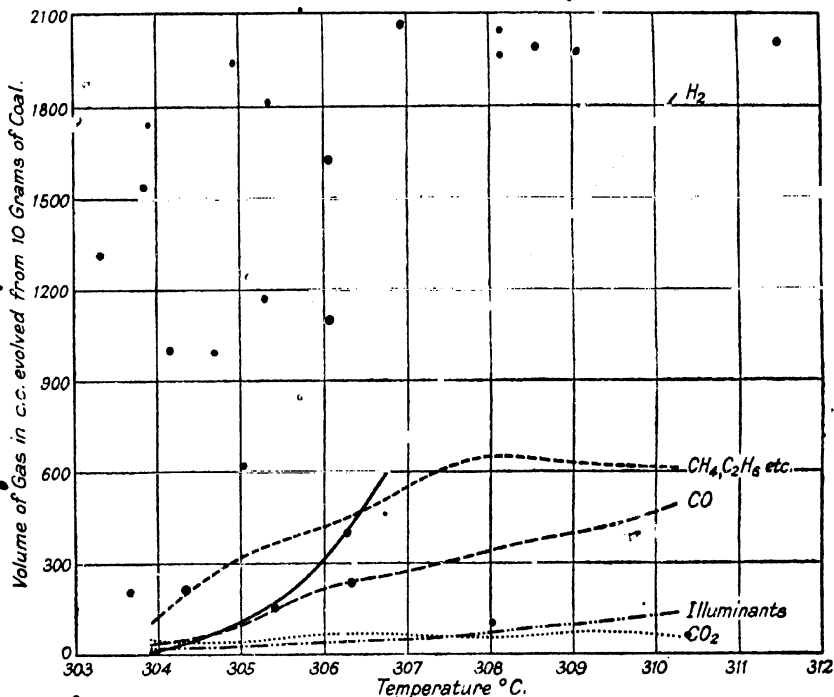
FIG. 4.—VOLUMES OF GASES EVOLVED FROM ILLINOIS COAL.

small quantity of air is contained in the tube before the experiment commences, the gas samples are to a small extent contaminated with air. The investigators point out that—

> "This explains the presence of oxygen, the high nitrogen figures, and probably accounts for a portion of the carbon dioxide, as oxygen is freely absorbed by coal at temperatures much below the point of decomposition. This absorption of oxygen makes it impossible to arrive at the exact composition of

the gas from the coal by deduction of the volume of air found by calculation, and the results are therefore reported in the form obtained from the analyses...."

The authors go on to state that—

"The analyses were carried out over mercury in a Bone and Wheeler apparatus, to which was fitted a copper oxide tube for the direct determination of the hydrogen. The proportions of the saturated hydrocarbons were calculated
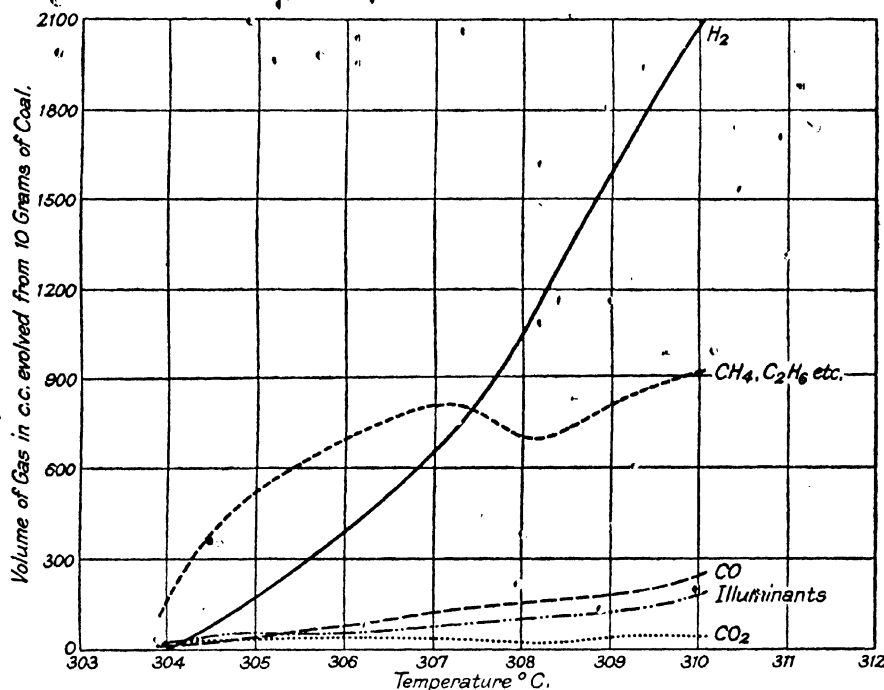
FIG. 5.—VOLUMES OF GASES EVOLVED FROM PENNSYLVANIA COAL.

from the results of the explosion analyses, assuming only methane and ethane to be present. Since it is practically certain that higher paraffins are present to some extent, this assumption is not justifiable ; but the close agreement between the volume of oxygen used up in the explosion and that theoretically required for the proportions of methane and ethane reported, indicates that propane and higher paraffins, if present, occur in relatively small proportion. . . .

"The proportion of nitrogen was arrived at by deducting from the total residual nitrogen the volume of the latter which had been added in the form

of air, to supply the oxygen necessary for the combustion of the hydrocarbons, instead of representing it as the difference between the 100 and the sum of the other constituents, as is customary. The variation of the total from 100 represents errors of analysis. The percentage of nitrogen reported in No. 5 is evidently low, as it falls somewhat short of the figure which is equivalent to 3 per cent of oxygen."

The principal data obtained by Gray and King are given in Table XXI. Their results, in common with those of the previous investigators, emphasise the high ratio of methane and higher paraffins in gas produced by low temperature carbonisation at temperatures of about 600° C.

Reference may also be made to a paper by Porter and Taylor[1] in which are shown the yields of typical coals between the temperatures of 250° and 450° C.

CRITICAL TEMPERATURES IN THE HEATING OF COAL

Somewhat similar yields of gases to those of Porter and Ovitz have been obtained by Burgess and Wheeler[2] on samples of bituminous, semi-bituminous and anthracite coals. It appears from their experiments that the occluded gases are evolved up to about 200° C., that the evolution of the combined water commences at 200° C. and that the decomposition of the coal substance commences at about 350° C. This is confirmed by some large-scale work at H.M. Fuel Research Station, when coal was raised to a temperature of 350° C. in a rotary heater.

Vignon[3] concludes that unsaturated hydrocarbons are evolved up to 600° C. but not beyond, and that methane and other paraffins form from 60 per cent to 80 per cent of the total yield of gas up to about 800° C.

It is clearly established that there is a definite critical temperature between 700° and 800° C., at which the evolution of hydrogen rapidly increases. This phenomenon has already been referred to (page 46, Chap. IV), and its exact significance need not here be discussed. It is generally agreed, however, that secondary reactions on a large scale begin at this temperature. The secondary reactions are due to the influence of heat upon the primary products of distillation previously formed, the tar being changed in character and diminished in quantity, and the gas being increased in volume but diminished in complexity and calorific value. This temperature of 700° C. may be considered the limit beyond which

[1] Porter and Taylor. "The Mode of Decomposition of Coal by Heat." Proc. Amer. Gas Ins., 9th Annual Meeting. Chem. Sec., 1914, 1–50.
[2] Burgess and Wheeler. "The Volatile Constituents of Coal." Trans. Chem. Soc., 1910, 97, 1917–35, and 1911, 99, 649–67. "The Distillation of Coal in a Vacuum." Trans. Chem. Soc., 1914, 105, 131–40.
[3] Vignon, L. "Distillation Fractionée de la Houille." Comptes Rendus Acad. Sci., 1912, 155. 1514–17.

TABLE XXI

Low Temperature Assay at 600°C. of Typical British Coals (Gray and King)

Coal	Proximate Analysis of Coal				Analysis of Gas (percentage volume).									Yield of Gas at 60° F. and 30″ saturated.	
	Moisture.	Organic Vol. Matter.	Fixed Carbon.	Ash.	CO_2.	$\dfrac{C_nH_{2n}}{C_nH_{2n:}}$	O_2.	CO.	H_2.	CH_4.	C_2H_6.	N_2.	Total.	C.Cs. per 10 gms. of coal.	S.G. (air = 1)
Coleorton House Coal	9.98	35.84	50.04	4.14	12.6	3.1	1.4	11.5	16.8	32.3	12.5	10.1	100.3	1004	0.76
Mapperley Small Nuts	7.23	34.74	51.11	6.92	13.7	3.1	1.6	11.5	19.8	31.5	11.2	7.6	100.0	1078	0.72
Rossington Main (2)	4.71	29.09	44.32	21.88	10.4	3.3	1.6	8.1	22.1	33.9	11.3	9.2	99.9	900	0.70
Wentworth Silkstone	1.32	36.31	58.55	3.82	5.6	3.4	1.9	5.7	23.0	39.4	12.3	8.6	99.9	1021	0.68
Newcastle Main, Manton Hard Steam	4.68	34.85	56.60	3.87	4.2	1.8	3.0	3.9	22.4	34.0	18.5	11.7	99.5	999	0.66
Dalton Main (" J ")	1.62	33.60	56.38	8.40	7.8	3.2	2.1	5.2	23.4	36.4	13.2	8.7	99.7	1051	0.66
Rossington Main (2)	6.37	33.40	56.26	3.97	—	—	—	—	—	—	—	—	—	1036	0.70
Bannockburn Hartley	0.92	25.67	71.54	1.87	3.35	3.05	1.1	3.7	26.35	40.05	14.1	7.8	99.5	1110	0.59
Nixon's Navigation	0.88	12.92	83.16	3.04	4.0	1.0	1.4	2.9	35.5	41.7	5.4	8.2	100.1	930	0.4x
Bwllfa Large Welsh	0.90	10.09	80.96	8.05	5.2	0.7	1.8	2.8	36.9	39.1	4.0	9.5	100.0	768	0.45

coal should not be heated in any process for low temperature carbonisation if the oils and gases are to possess the properties associated with the primary products of carbonisation.

THE EFFECT OF THE CARBONISING TEMPERATURE ON THE QUALITY OF THE COKE

The effect of the temperature of carbonisation on the quality of the resulting coke is very marked for wide temperature ranges. The structure of the coke resulting from the carbonisation of the same coal at various temperatures has been examined by Cobb and Greenwood.[1] In particular the specific gravity, porosity and weight of the resulting samples of coke were determined. The temperatures employed were 550° C., 850° C. and 1100° C., corresponding to low temperature, gasworks and coke-oven practice respectively. The original coal had a specific gravity of 1·27. The specific gravity and porosity, as calculated from the ratio of actual coke volumes and interspaces, of the resulting coke were as follows :—

		Temperature	Specific Gravity.	Porosity.
A	. . .	550° C.	1·59	44·7
B	. . .	850° C.	1·87	52·5
C	. . .	1100° C.	1·87	48

The differences are summarised by the authors of the paper as follows :—

(1) The total volume of coke A was 3 per cent greater than that of the original coal. Such swelling is the cause of much difficulty in low temperature carbonisation.

(2) Coke B was much harder than coke A, and the changes in weight, porosity, etc., show that this hardening was accompanied by a thinning of the cell walls.

(3) Coke C had similar properties to those of a commercial sample of metallurgical coke. The hardening in this case was accompanied by a thickening of the cell walls. This circumstance is regarded as one main difference between gas coke and hard metallurgical coke.

THE TEMPERATURE OF CARBONISATION

From the data hitherto published of the results of laboratory experiments it would appear that there is not yet definitely established a temperature lying between 350° and 750° C., for which it may be said that the yields of the products of distillation are an optimum. It is possible, however, that on a commercial plant for low temperature carbonisation the working range of temperature may fall within

[1] Cobb and Greenwood. "The Structure of Coke." Journ. Soc. Chem. Ind., March, 1922.

narrow limits. The value of the gas, tar and coke must then be measured in terms of the quantity and quality from an economic standpoint, while the practical difficulties of maintenance and upkeep of the plant must also be borne in mind. The effect of heat on the quality and strength of iron and steel at high temperatures must in particular be taken into account. It will possibly be found that a temperature in the neighbourhood of 600° C. is, on the whole, most suitable for the majority of commercial plants, though it may be desirable to heat certain coals to a higher temperature, say 650° C., in order to assist in the production of a compact coke.

B. THE TIME REQUIRED FOR CARBONISATION

THE TIME ELEMENTS IN THE CARBONISATION PROCESS

It is essential from a commercial standpoint to have reliable information on the time required for the carbonisation of coal. The conditions under which laboratory experiments are performed are, however, so different from those which occur on any full-scale plant that it is very difficult to draw any conclusions of value for commercial application. Practical aspects of the problem of heating the material will be discussed in a later chapter. In this section the matter will be considered in a preliminary manner from the results of small- and medium-scale experimental work.

Coal is a poor conductor of heat, and the time of carbonisation is influenced very largely by the method of heating, the shape of the retort, the thickness of the charge, the size of the particles, the closeness of packing, the facilities for the escape of the vapours and many other important factors. Whereas detached particles of coal when exposed to radiant heat can be carbonised in a few seconds, it has been found that the carbonisation of 20 grammes of finely ground coal in the laboratory assay apparatus of the Fuel Research Board is only completed when a temperature of 600° C. has been maintained for at least twenty minutes. Apart, therefore, from the time required to transfer the heat from the heating medium to the charge, which is influenced by the method of heating, size of particles, etc., it would appear that there is a further time-element, due to the necessity for maintaining the coal at the maximum temperature before carbonisation is complete. It is scarcely to be expected that the varied and complex reactions which occur in a mass of coal heated to a temperature of, say, 600° C. can be completed immediately that temperature is attained, and an interval of time must elapse before the gases and volatile products can be removed from the charge. Both the variables, rate of heating and rate of completion of the carbonising reactions are dependent, among other factors, upon the ratio of the mass to the surface of the charge.

THE EFFECT OF THE TIME-ELEMENT ON THE YIELD OF GAS

In connexion with the effect of the time-element on the yield of gases, results of interesting experiments on a works scale are given in the Report of the Fuel Research Board.[1] In these experiments the variations in the rate of evolution, in the calorific value and the density of the gas at different stages of the carbonising period were determined at a constant retort temperature of 600° C.

The graphs of typical results on a time basis are shown in Figs. 6, 7, 8 and 9. It will be seen that there is an initial rapid evolution of inert gas, probably chiefly of steam and carbon dioxide, although some of the effect observed is due to expansion
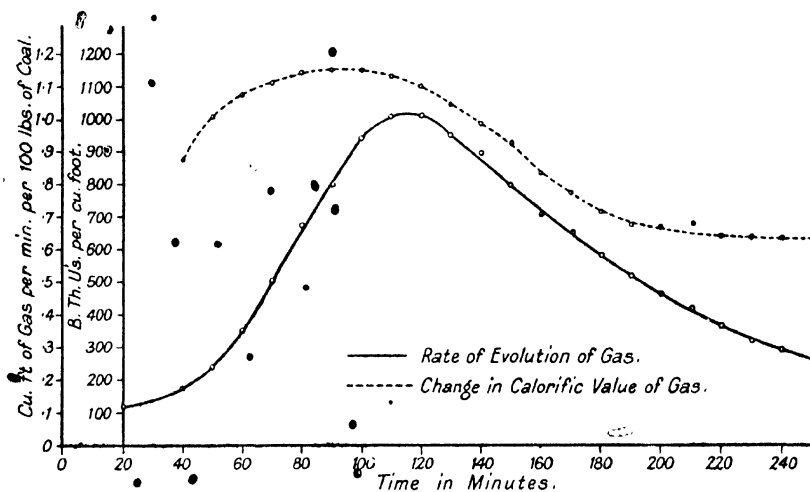
FIG. 6.—CARBONISATION IN HORIZONTAL STEEL RETORTS OF LANGLEY BRIGHTS COAL.

of cold air which is admitted with the coal charge. The rate of evolution then drops to a minimum at about 30 minutes, but later steadily rises to a maximum at about 120 minutes, after charging, but is sustained for about 20 minutes only. The peak in the curve of calorific value occurs at about 1½ hours after charging, where it reaches a value of about 1200 B.Th.U. ; but it will be noticed that, after about 2½ hours, it drops very rapidly to about 700 B.Th.U. and less. The specific gravity starts at a maximum value, and drops steadily to a fairly constant figure at 2½ hours and onwards. It thus follows that, from the standpoint of the yield of gas, there is no advantage to be gained in " stewing " the coal for an indefinite period, since

[1] Fuel Research Board. Report for the years 1920-1. Second section : Low Temperature Carbonisation." (H.M. Stationery Office.)

the increased quantity of gas to be obtained is small. Fig. 10 shows a typical gas evolution curve for the laboratory assay apparatus for a similar coal to that from which Fig. 8 has been obtained. It will be seen that these curves present a remarkable similarity in shape.

THE EFFECT OF THE TIME-ELEMENT ON THE STRUCTURE OF THE COKE

The effect of the time-element on the structure of the coke is of considerable importance. This is discussed very fully in the report already quoted, and to which further reference should be ma le. It has already been pointed out that small
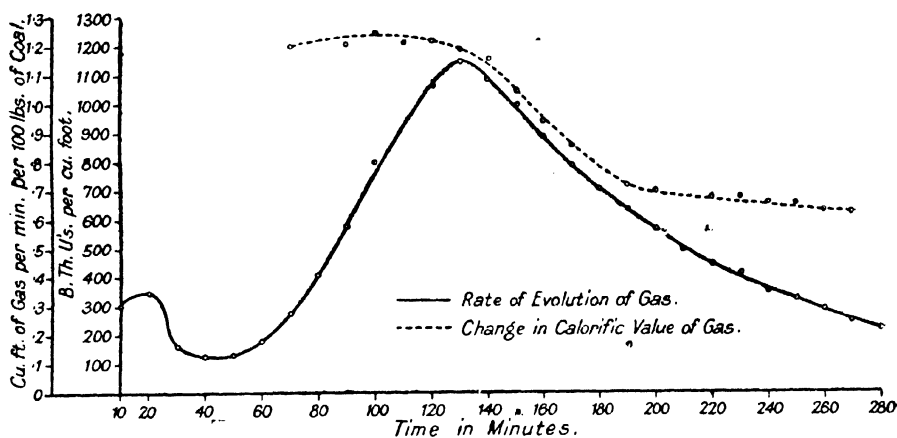
FIG. 7.—CARBONISATION IN HORIZONTAL STEEL RETORTS OF DALTON MAIN COAL (SAMPLE 1).

detached particles of coal can be carbonised by radiant heat in a few seconds, but that 20 grammes of finely ground coal are only carbonised in the assay apparatus when a temperature of 600° C. is maintained for at least twenty minutes. Thus, as the ratio of surface to mass is decreased, the "time-element" in carbonisation becomes important. This has clearly an important bearing on the problem of carbonising material in thin layers.

On the other hand, any attempt to take advantage of the rapid action of radiant heat introduces certain undesirable complications when the coke is required for domestic purposes. In the earlier experimental work at H.M. Fuel Research Station, when coal was carbonised in shallow trays in the plant described on page 220, it was found that the effect of the radiant heat from the crown of the steel retort was to carbonise the surface layers so rapidly that little or no general

fusion had taken place. This effect was so great that, in a layer of material one inch thick, the strength of the final coke was adversely affected. Although the disadvantage was obviated by placing a steel plate on the top of the carbonising mass, this solution of the difficulty was, of course, undesirable from a manipulative standpoint. It should be noted in passing that the reduction of the " time-element " by the utilisation of radiant heat would be of considerable advantage if the resulting fuel could be used either in the form of powder or as the material in a briquetting process.

In consequence of this experience a thicker layer of material was used and the
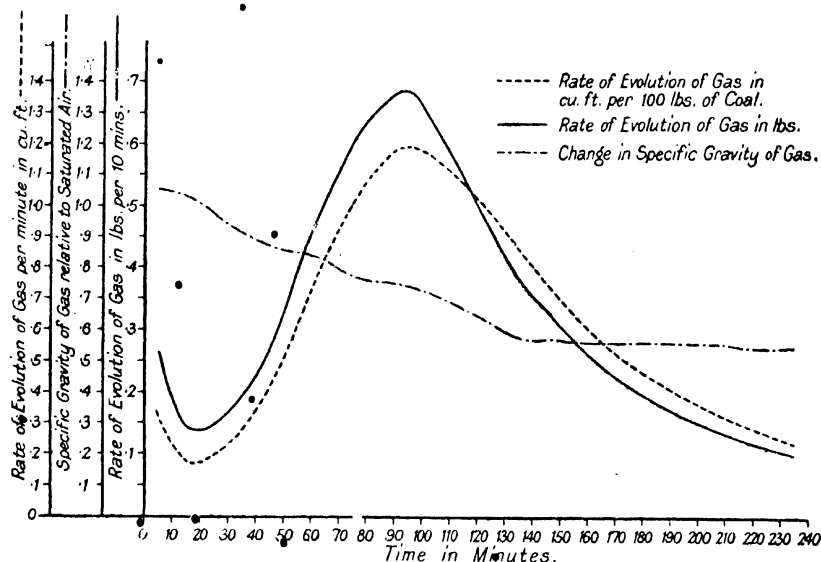
FIG. 8.—CARBONISATION IN HORIZONTAL STEEL RETORTS OF DALTON MAIN COAL (SAMPLE 2).

carbonising trays were divided into square cells by means of grids made of half-inch steel strips, in order to assist in heat distribution by conduction and so compensate for the increased thickness of the coal mass. Blocks of fuel were thus prepared, and those of three-inch cube were found the most satisfactory. Five of the six sides of the blocks being in contact with the steel surfaces were smooth and glazed, the sixth side being rough. The high heat conductivity of the steel facilitated the flow of heat to the material in the centre, and so reduced the time of carbonisation, and the blocks were found to be of uniform structure. Further useful information could be deduced from the fact that under these conditions the presence of a layer of finely powdered coke on the free surface gave evidence when the mixture of coal

employed was deficient in caking power. Such blocks were adopted as the standard form of fuel produced by this particular process.

The effect of the passage of volatile gases through the mass of coal is of import-ance, especially during the cementation period, i.e. during the period when the temperature lies between 350° C. and 450° C.

During this period the material is very resistant to the passage of gas, and, while the effect of this may not be pronounced when the material is treated in thin layers, the effect when it is heated more or less in bulk, as in certain
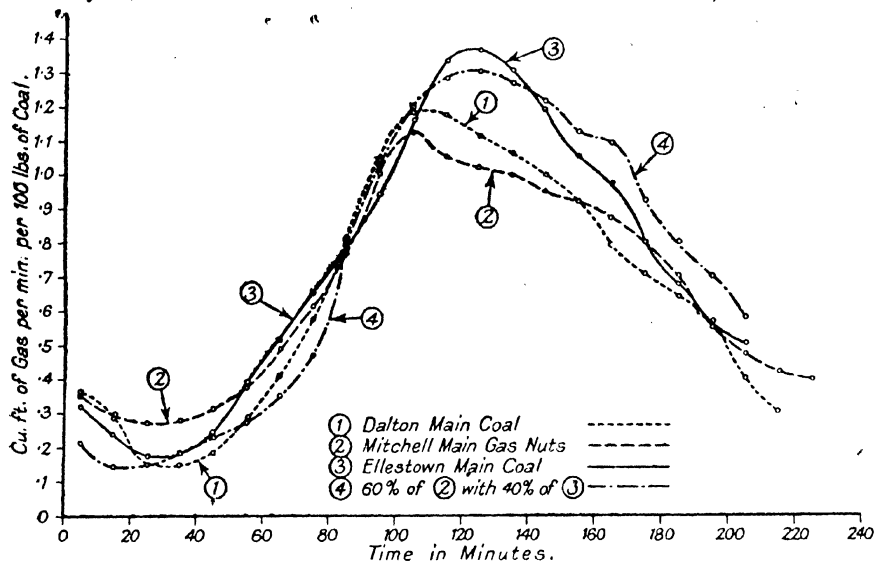
FIG. 9.—COMPARISON OF RATE OF EVOLUTION OF GAS DURING CARBONISATION AT 600° C. OF VARIOUS COALS IN HORIZONTAL STEEL RETORTS.

types of vertical retorts, may be very marked. Consider, for example, the material in a vertical retort which is externally heated. Owing to the poor heat conductivity of coal, the zones of temperature from the walls inward may be taken to vary somewhat, as shown in Fig. 11. The material in zone A being above the cementation temperature has solidified and will be pervious to the gas. The material in zone B being at the cementation temperature and in a plastic condition prevents the flow of gas from C to A or A to C. As the heat is further transmitted to the coal, the plastic envelope travels towards the centre of the core. The fusing of the coal tends to resist the flow of the gases, forcing a large proportion of them to escape through the outer layers of material at the higher temperatures. In order to prevent the pressure within the retort rising unduly, and to facilitate the transfer

of heat to the interior by the medium of the carbonisation gases, it is desirable in such cases to use screened coal of uniform size, or to stop the expansion by other means, such as blending.
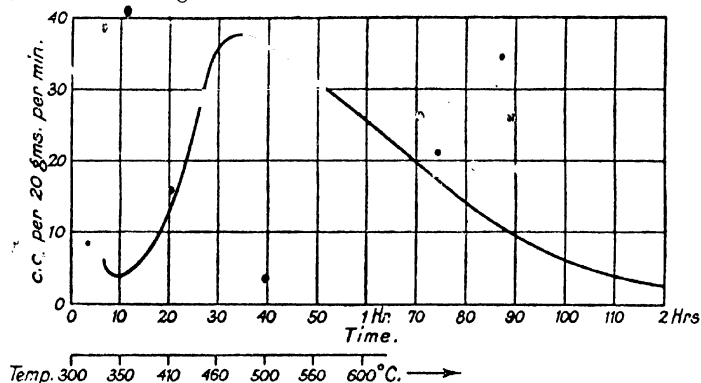
FIG. 10.—GAS EVOLUTION CURVE IN LOW TEMPERATURE ASSAY APPARATUS OF DALTON MAIN COAL.

The effect of these aspects of the time-element on the carbonisation of coal at low temperatures is thus of great importance, and should be closely studied by those interested in the design of a full-scale plant.

THE EFFECT OF·THE TIME-ELEMENT ON THE YIELD OF TAR

The changes likely to be brought about by modifications in the elements of carbonisation are dealt with more fully in Chapter XIII. It is, however, desirable to note at this stage that the effect of the time-element on the yield and quality
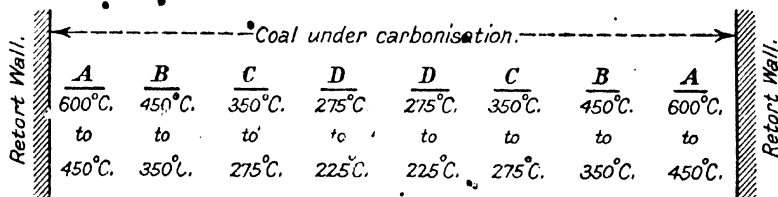
FIG. 11.—ZONES OF TEMPERATURE IN EXTERNALLY HEATED VERTICAL RETORTS.

of tar is roughly in conformity with its effect on the gas. The influence of time is bound up with the speed at which the tar vapours are removed from the heated zones ; thus, if their removal be very slow, the tar vapours will experience secondary decomposition, with the production of lighter fractions, gas, carbon and pitch. The yield of oils will be decreased, and the character of the bodies formed begins to change from paraffinoid to benzenoid structure.

Time has also another effect quite apart from changes due to prolonged heating of the oil which has already been formed and vaporised. During the carbonisation of coal there will be a period when tar formation is at a maximum rate, after which it falls off in somewhat the same manner as we have already seen in the case of the gas. That is to say, oils will still continue to be given off, but in rapidly diminishing quantities. As in the case of the gas generated, a moment will be reached when tar formation, while still continuing, will be so small as to render it uneconomical to prolong the carbonisation any further for oil production alone. In most practical cases, however, carbonisation is carried out at such a temperature that the evolution of tar is practically complete before the rate of evolution of gas has fallen to such a figure as would render it uneconomical to continue carbonisation.

C. THE AMOUNT OF HEAT REQUIRED FOR CARBONISATION
EXOTHERMIC AND ENDOTHERMIC REACTIONS

The amount of heat required to produce the necessary chemical and physical changes for the carbonisation of coal is not large, but its economic importance varies according to the method of carbonisation adopted.

The problem is one of extraordinary complexity, since the original coal is a heterogeneous material containing a large number of chemical entities which react together under the influence of heat to produce solids, liquids and gases of equal complexity. When a reaction takes place between two chemical compounds, heat is either absorbed or evolved. If the reaction is accompanied by an absorption of heat it is said to be endothermic, while if heat is given off the reaction is termed exothermic. Thus, if carbon is burnt in oxygen, according to the reaction $C + O_2 = CO_2 + 97,000$ calories, heat is given off to the extent of 97,000 calories per gramme molecule of carbon dioxide formed. The reaction is therefore accompanied by a rise in temperature of the product, and is exothermic. On the other hand, if carbon be made to combine with hydrogen according to the reaction $2C + H_2 = C_2H_2 - 48,000$ calories, heat is absorbed to the extent of 48,000 calories per gramme molecule of acetylene formed. The reaction can only take place if heat is supplied, and is therefore endothermic. This heat apparently disappears as such, but still remains as energy in the substance or substances formed, and this energy can be regenerated into heat units by suitable means.

THERMAL REACTIONS IN COAL AT HIGH TEMPERATURES

In the carbonisation of coal we find exothermic and endothermic reactions taking place simultaneously. These tend to balance each other thermally, and the net result is only exothermic or endothermic to a slight degree. Work on this

subject has been carried out by Mahler[1] in 1891, Euchène[2] in 1900, Constam and Kolbe[3] and Schlapfer[4] in 1908 and 1909, and Hollings and Cobb[5] in 1914 and 1915.

Mahler determined for a certain type of coal in his bomb calorimeter the heat of combustion of the coal, and also that of all the products of carbonisation, finding that the latter was less than the former by some 459 B.Th.U. per lb. of coal, or about $2\frac{1}{2}$ per cent of the heat in the original coal.

Euchène compared the heat of formation of the distillation products with the heat balance of a retort setting, and for coal of a similar ultimate chemical analysis to that used by Mahler found an evolution of heat during the reactions of 114 B.Th.U. per lb., or say 0·6 per cent. In his paper Euchène discussed observations made with three types of coal which differed in their content of oxygen and also in their volatile matter, all of which showed a liberation of heat during the distillation. The amounts apparently liberated in his experiments are, however, very small (always under 1 per cent), and are therefore of an order comparable with the probable percentage error of his experiments.

Constam and Kolbe, using the method adopted by Mahler, worked on typical English coals, and obtained figures for an evolution of heat varying between 6 per cent for a Nottinghamshire coal and 2·1 per cent for a Welsh steam coal.

THERMAL REACTIONS IN COAL AT LOW TEMPERATURES

The whole of the work mentioned above refers to the overall evolution of heat during a process where coal is distilled by subjecting it to temperatures which increase from atmospheric at the commencement to the temperature associated with high temperature carbonisation. Little work of a quantitative nature has up to the present been attempted where the maximum temperature has been limited to those associated with low temperature carbonisation. Hollings and Cobb[6] have, however, compared the temperature changes which occur in a sample of coal when it is heated and distilled by a definite inflow of heat with those which occur during the heating of a similar sample of inert material by a similar inflow. In this way they have succeeded in tracing the course of the exothermicity or endothermicity of the reactions which take place at the different temperatures. Bituminous coals, anthracites, cannel and cellulose were examined, and they were able to detect

[1] Mahler, A. "Sur la Distillation de l'Houille." *Comptes Rendus*, 1891, **113**, 862.
[2] Euchène. "Thermic Reactions in the Distillation of Coal." *Journ. Gas Lighting*, **76**, 1080, 1900.
[3] Constam and Kolbe. "Researches on the Carbonisation of Typical English Coals." *Journ. of Gas Lighting*, **107**, 696, 1909.
[4] Schlapfer. "Study of the Carbonisation of the Chief Types of Coal." *Journ. of Gas Lighting*, **103**, 382, 1908, 1909.
[5] Hollings and Cobb. "A Thermal Study of the Carbonisation Process." *Gas Journ.*, 1914, **126**, 917; *Trans. Chem. Soc.*, 1915, **107**, 1106; *Fuel in Science and Practice*, Nov., 1923.
[6] Hollings and Cobb. "A Thermal Study of the Carbonisation Process." *Gas Journ.*, **126**, 917, 1914; *Trans. Chem. Soc.*, 1915, **107**, 1106; *Fuel in Science and Practice*, Nov., 1923.

"a number of fairly well-defined stages during which exothermic or endothermic reactions predominate." In their summary Hollings and Cobb state : " In the case of the Monckton coal used, the distillation, so far as it is revealed by thermal changes, commences at 250° C., and endothermic reactions predominate up to 410° C. This is followed by a short exothermic stage between 410° C. and 470° C., and a second endothermic period between 470° C. and 610° C. The exothermic stage above 610° C. is always very pronounced, and is interrupted between 750° C. and 800° C. The probability that exothermic reactions continue to some higher temperature has already been discussed."

It will be seen then that, although the overall reactions associated with high temperature carbonisation appear to be exothermal, it does not follow that this will apply to the results of a low temperature carbonisation method. If the temperature be limited to 600° C. in the case of the coal used by Hollings and Cobb, we have endothermic reactions commencing at 250° C. and continuing until 610° C., with a break between 410° C. and 470° C., during which there is a short exothermic stage. For this coal, at any rate, it is possible that the overall reactions during low temperature carbonisation may be either neutral or slightly endothermic. It may, however, be pointed out that in most of the coals which have been tried under low temperature conditions at H.M. Fuel Research Station the potential heat in the products after cooling to datum temperature has been less than the sum of the potential heat in the raw coal and the sensible and latent heat of any steam used. It must not be forgotten, however, that the results of such determinations include the sum of all the errors made in the necessary measurements.

In large-scale work, where experimental conditions are necessarily more crude, the effect of heat loss by radiation, etc., which cannot be directly measured is so great as to obscure the relatively smaller effect of the heat balance of the reactions which take place in the coal substance.

THE HEAT BALANCE SHEET IN PRACTICE

Consider the heat balance sheet for coal carbonised in a large-scale plant at any given temperature. On one side we have all the heat entering the system, this being the sum of the potential heat of the coal, the heat both sensible and latent of any steam used, and the heat supplied by the combustion of the fuel burnt in the setting. On the other side we have the potential heat in the products —coke, oil, gas and liquor—after they have been cooled to the datum temperature, the heat both sensible and latent of these products as they are removed from the retorts and which has been mainly abstracted in the condensing and cooling plant, the heat present in the products of combustion of the fuel burnt as they leave the setting, and the heat lost from the setting by radiation, conduction and convection to surrounding bodies.

If all of these were capable of accurate measurement the two sides of the account would balance. In practice even with the most careful measurements a balancing figure would be required which would represent the algebraic sum of the errors made in the separate measurements. While experiments have been attempted in high temperature practice with an approximation to the degree of elaboration quoted above, there has as yet been no such work carried out for low temperature processes.

If again we consider the heat balance sheet for the retorts only, we have on one side the potential heat of the coal, the heat both sensible and latent of any steam used and the heat which has passed through the walls of the retort from the heating chambers. On the other side we have the potential heat in the products of carbonisation after cooling to datum temperature and the heat both sensible and latent of these products as they leave the retort.

It is often necessary when considering the design or application of any type of retort to form some opinion as to the amount of heat which it will be necessary to supply to the setting in the form of fuel burnt. On one side of the heat account for the combustion chambers we have the potential heat supplied to the setting whilst on the other side we have the heat passing through the retort wall to the charge, the heat lost in radiation, conduction and convection and the heat both sensible and latent in the products of combustion as they leave the setting. Of these an estimate of the loss by radiation, etc., may be made from analogies with plant of a similar nature.

With regard to the heat which must be passed through the retort walls to the charge a consideration of the retort heat balance will show that this can be obtained by subtracting the sum of the potential heat in the coal and the sensible and latent heat of any steam coal used from the sum of the potential heat in the products after cooling to datum temperature and the sensible and latent heat of the products of carbonisation as they leave the retort.

In practice the potential heat in the products is less than that in the coal and steam charged, and the following examples of the difference as obtained from two types of plant are quoted from experimental work at H.M. Fuel Research Station: —

Horizontal Retorts (described on page 220). 5-8 per cent of potential heat in coal.

Vertical Retorts (Technical Paper No. 7,.Fuel Research Board). 1·3-3·7 per cent of the sum of potential heat in coal and in steam supplied.

If, then, figures of this magnitude are subtracted from an estimate of the sensible and latent heat of the products as they leave the retorts an idea can be obtained of the heat which must be supplied through the retort wall in order to effect the process.

CHAPTER VIII

TECHNICAL PROBLEMS—METHODS OF HEATING THE MATERIAL

INTRODUCTORY

TURNING now from the study of the chemical and physical aspects of the subject, it is desirable to discuss certain general principles affecting the design of plant required for low temperature carbonisation before attempting to describe some typical retorts already tried or proposed. In this way the possibilities and weaknesses of these retorts may perhaps be the better appreciated, and the prospects as well as the difficulties of the different processes may be the more easily appraised.

Retorting systems may be broadly divided into three main groups. In the first, the crushed coal is exposed in thin layers in a gas-tight retort or chamber to the action of heat, which is passed through the walls of the retort from an outside oven or combustion chamber, and is distributed to and through the mass by conduction, radiation and convection. The layer of coal is maintained in repose during the period of carbonisation—that is to say, it is not mechanically turned over for the exposure of new surfaces to the action of the heat.

In the second, the crushed coal is exposed to heat in an externally heated gastight chamber in which it is mechanically stirred and turned over, so that fresh surfaces are exposed to the radiated, convected and conducted heat from the walls of the chamber. Retorts of this type may be horizontal, vertical or inclined, and the turning over of the coal may be effected by the action of gravity or by the movements of mechanical apparatus placed within the retort for the purpose.

In the third, the crushed coal is exposed to the action of heat derived from the sensible heat of a large volume of producer gas, superheated steam or some inert substance which is passed over and through the mass of broken coal. If producer gas is used, it is either generated by the combustion in a mixture of air and steam of a proportion of the coke which results from carbonisation within the producer itself, or is generated in a separate gas producer fed with an independent supply of coke.

Differentiating by the method of heating, these groups form two classes. Groups one and two fall together in an externally heated class of retort, and group three forms an internally heated class. Even with this classification there is a certain amount of overlapping, since retorts have been suggested which combine external with internal heating in one apparatus.

The methods of supplying heat for the carbonising process have a very important bearing not only in determining the design of the retort and in modifying the products of carbonisation, but also possibly in fixing a maximum working limit to the thermal efficiency of the process.

In the case of the externally heated retorts the material to be carbonised is

supplied with heat through the walls of the retort, i.e. the hot flue gases or the flames from the source of heat do not come in contact with the charge within the retort. In the case of the internally heated retorts the material to be carbonised is heated by direct contact, i.e. by passing the hot gases or other substances into the retort and bringing them into intimate contact with the charge. The differences brought about by these alternative methods are so great that it is essential to discuss them in some detail. Before doing so, however, it will be desirable to consider some points affecting the size of material employed for processes of low temperature carbonisation.

SIZE OF MATERIAL

There is great variation in the sizes of material which inventors have attempted to use in the different processes of low temperature carbonisation put forward by them. The maximum size for certain proposed installations is given as $2\frac{1}{2}$ inches, but for others only $\frac{1}{4}$ inch or even less. In one process, which at the present time gives indications of great promise, the most desirable size is given as double or treble nut, the finer sizes being screened out. In most cases, however, especially when blending is adopted, it is advantageous to have the material crushed, so that the pieces have a large surface in proportion to their volume. The weight of a given volume of the material is increased by its fine division, the mixing can be made more intimate, and the resultant coke is improved both in density and robustness.

Many experiments have been carried out at H.M. Fuel Research Station to determine the influence of the size of material on the rate of its carbonisation in trays in externally heated retorts, and on the robustness of the resulting coke. It was found that coal crushed and left on a 4-mesh sieve did not give a satisfactory coherent coke. It could be broken down very readily, and would not be suitable for transport. On the other hand, by leaving the dust and finer particles in the crushed coal, the resulting coke was much more robust. It was found that the thermal conductivity of the material was also improved. In the experiments on low temperature carbonisation described in the Report of the Fuel Research Board[1] the standard of fineness was taken to be that of crushed material which had been passed through a disintegrator of the double cage type. Table XXII taken from that Report gives the typical proportions of particles of various sizes after disintegration in this machine.

The best degree of fineness for material to be carbonised in certain types of continuous vertical retorts is not, however, the same as that for material carbonised in trays in a horizontal retort. Similar experiments to determine the best size of material for carbonisation at low temperatures in one type of externally heated

* [1] Report of the Fuel Research Board for the years 1920-1921 ; Second Section "Low Temperature Carbonisation." (H.M. Stationery Office.)

vertical retort, where gravity is relied upon to secure the continuity of the process, are described in a further Report of the Board.[1] Two retorts were charged with disintegrated coal and two with uncrushed coal (nut size). The uncrushed coal gave good results, whereas the disintegrated coal did not apparently move uniformly down the retort.

TABLE XXII

TYPICAL PROPORTION OF PARTICLES OF VARIOUS SIZES USED IN EXPERIMENTS AT H.M. FUEL RESEARCH STATION

Passing 4 mesh and remaining on 10 mesh sieve . . . . 24 per cent.
,, 10 ,, ,, ,, 20 ,, ... 27 ,,
,, 20 ,, ,, ,, 30 ,, . . . 14 ,,
,, 30 ,, ,, ,, 40 ,, ... 7 ,,
,, 40 ,, ,, ,, 50 ,, ... 6 ,,
,, 50 ,, ,, ,, 60 ,, ... 4 ,,
,, 60 ,, ,, ,, 100 ,, ... 5 ,,
,, 100 ,, ,, ,, — ,, ... 13 ,,

The weight per cubic foot of Dalton Main coal so treated was 49 lb.

but formed a hard crust, which moved slowly down the walls and allowed partially carbonised coal to pass more rapidly down the centre. The size of coal successfully used in the experiments described in this Report was :—

> Amount remaining on 1-inch sieve . . . 20·4 per cent
> ,, ,, ,, ½-inch ,, . . . 73·8 ,,
> ,, ,, ,, ¼-inch ,, . . . 89·5 ,,
> passing ¼-inch sieve . . . 10·5 ,,

It should be noted that, in the vertical retorts used in the Scottish shale industry, the shale is passed through in lumps of fairly definite size. In such cases the openness of the material undergoing carbonisation allows the free play of convection currents, and so facilitates the passing of heat to the interior of the charge, with the result that a more uniform distillation is effected. It will be seen then that the results of experiments with coal in vertical retorts at H.M. Fuel Research Station are quite in keeping with the long experience of the shale oil industry.

Experiments on the effect of the degree of fineness of coal in high temperature practice have also been made by Mr. T. Biddulph Smith.[2] Material of the three degrees of fineness given in Table XXIII was carbonised in a coke-oven under

[1] Fuel Research Board Techr Paper No. 7. "Preliminary experiments in the Low Temperature Carbonisation of Coal in Vertical Retorts." (H.M. Stationery Office.)
[2] Biddulph-Smith, T. "Effect of some Physical Conditions during Carbonisation of Coal upon the Quality of Coke Produced." Coke Oven Managers' Assn., 19th April, 1922.

similar conditions, and the strength of the resulting coke measured. These readings show that the gain in the strength of the coke obtained from finely ground material is very great, although in this case it must be pointed out that the coke, being made at a high temperature, was for use in a blast-furnace, and not in a domestic fire-place.

TABLE XXIII

STRENGTH OF COKE OF VARYING DEGREES OF FINENESS (BIDDULPH-SMITH)

	No. 1. Per Cent.	No. 2. Per Cent.	No. 3. Per Cent.
Between 1 in. and $\frac{1}{4}$ in.	Nil.	Nil.	36·5
Between $\frac{1}{4}$ in. and $\frac{1}{10}$ in.	Nil.	3·7	27·0
Between $\frac{1}{10}$ in. and $\frac{1}{30}$ in.	Nil.	51·8	31·3
Between $\frac{1}{30}$ in. and $\frac{1}{60}$ in.	Nil.	29·5	2·9
Between $\frac{1}{60}$ in. and $\frac{1}{90}$ in.	Nil.	10·0	1·8
Under $\frac{1}{90}$ in.	100	5·0	0·5
	100	100	100
Crushing stress of the resulting coke —lb. per sq. in.	1003	595	212

The effect of the subdivision of the material on the homogeneity and porosity of the coke is of great importance. Reference may be made in this connexion to the results obtained by Sutcliffe and Evans on the briquetting and carbonisation of pulverised fuel. In the process developed by these inventors finely ground coal is briquetted cold by subjecting it without any added binder to a pressure of 10 tons per square inch. The coke obtained on carbonising the briquettes is more dense than ordinary gas or metallurgical coke, and its homogeneity is remarkably good.

Such a treatment before carbonisation renders it possible to combine the advantages of hardness and closeness of texture associated with the use of finely divided material with those advantages more usually associated with the use of material in the lump form.

These do not, however, exhaust the considerations which tend to determine the size of the material for particular processes. The cost of grinding the material into fine particles is of great importance. Moreover, in certain cases—for internally heated vertical retorts, for example—there must not be too large a proportion of "fines," otherwise the material tends to pack in the retort, and mechanical means may have to be adopted to secure the continuity of the process. In addition, the evolved gases cannot pass freely through the retort and tend to form channel ways through the material, thus preventing the uniform coking of the charge. It must, however, be counted a disadvantage in connexion with any process if it rejects

the fine coal, since the working costs are simultaneously increased by the non-utilisation of a portion of the fuel available.

Whether the retorts are externally or internally heated, the heating effect of the gases evolved from the material is of some importance in reducing the time of carbonisation. In vertical retorts part of the carbonisation is effected by the hot gases from the lower parts passing up through the incoming charge. This effect is perhaps more noticeable with those coals which produce large quantities of gas. It is desirable that the material should not be in such a condition that on carbonisation the gases of distillation are restricted in their freedom in passing through the material.

A. EXTERNAL HEATING

PROBLEMS OF EXTERNAL HEATING

When the heating is effected externally, the hot flue gases are circulated round the body of the retort, and the heat is transferred through the walls to the charge within. The retorts used in the gas industry are of this type, and it is probably on this account that the great majority of the proposed retorts for low temperature carbonisation are in the same class. It should be added that the minority, the internally heated type of retort, form a small but important class.

TRANSFERENCE OF HEAT

In the following discussion on the question of transference of heat to the charge, the influence of the thickness and of the material of the retort walls will not be taken into consideration. There is in every externally heated retort a certain temperature drop between the outside and the inside surfaces of the retort, but reference will be made to this in a later section. It is desired at this stage to discuss the heat transference from the inner surface of the retort to the material which is being carbonised. In this transference, conduction, convection and radiation play important parts.

It is known that coal is a very poor conductor of heat, though it is difficult to give precise data for its conductivity or non-conductivity. The thermal conductivity of metals and some other substances can be fairly accurately determined, but that of coal and carbonaceous materials in general cannot be precisely obtained. The physical characteristics and chemical composition of coals vary considerably ; and, more important still, chemical and physical changes occur at even relatively low temperatures.

Apart from these variable features, the coal in practice is broken into small pieces before being fed into the retort, and the unknown variations in the interstitial spaces are a further difficulty in determining precise data on the rate of

heat transference through the coal. The influence of tightness of packing is well illustrated by some experiments made by one of the authors on the apparent conductivity of slag wool contained between two plates of sheet iron. If the wool were packed solidly, so that its apparent density approached that of the solid slag of which the filaments of the wool are composed, we should expect the conductivity to approach that of slag, and therefore to be high. Again, if the space between the two plates contained no slag wool, there would be free play for convection currents, and the flow of heat from plate to plate would again be high. The effect of the wool (the material of which is an excellent conductor of heat), is to prevent the free play of convection currents, and so we get little heat passing. Fig. 12
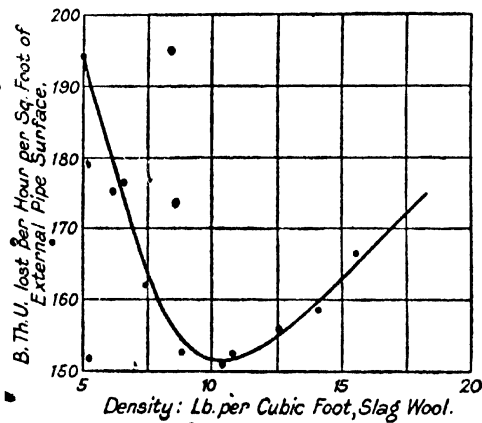
FIG. 12.—EFFECT OF DENSITY OF SLAG WOOL STEAM-PIPE COVERING ON HEAT LOSS.

illustrates the effect of varying the density of packing. On the left-hand side of the diagram the packing is light, and convection currents are insufficiently repressed. On the right-hand side of the diagram there is little convection, but the heat is transferred readily by conduction through the material of the wool. At the centre we find a point of minimum apparent conductivity corresponding to a density of packing of 10 lb. per cu. ft.

A similar effect is to be expected, and indeed can be observed, in the case of coal of varying fineness, but exact figures are not at present available. Calculations on the flow of heat through the coal are accordingly based on works experience rather than on laboratory experiments. It is found that the time of carbonisation of a charge is affected not only by the mode of heating, but also by the size of the material and the thickness of the layer employed.

In this connexion some experiments are published by Mr. Nielsen[1] which illustrate the slowness with which heat passes to the interior of a fairly tightly packed charge. A slab of coke from coal slack, about 3 inches thick, carbonised by being heated on one side only for about six hours in an externally heated vertical retort at a temperature of about 600° C., was carefully divided into four sections, parallel to the retort wall. The percentage of volatile matter in each section was determined separately, and the results are shown plotted in Fig. 13. It will be seen that the volatile matter varies from 10½ per cent near the retort wall to about
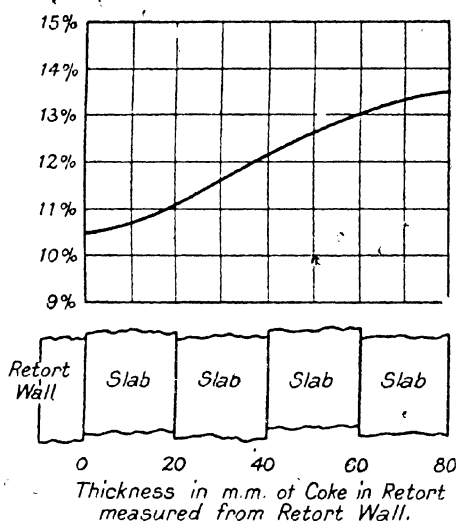
FIG. 13.—PERCENTAGE OF VOLATILE MATTER LEFT IN
LOW TEMPERATURE COKE (EXTERNALLY HEATED
RETORT).

13½ per cent at the centre of the charge. That is, in a given time the material near the retort walls is carbonised more than the material at the centre of the charge. This lack of uniformity in the carbonisation must be attributed mainly to the slow rate of heat transference across the coal.

A report of the Fuel Research Board[2] recently published gives an account of some interesting observations of the temperature conditions in a Glover-West retort in which experiments in carbonisation at low temperatures were being conducted. The charge consisted of a mixture of a strongly swelling coal (Mitchell Main) with a non-swelling coal (Ellistown Main) in the proportion of 60 per cent

¹ Nielsen, Harald, " Producer Gas for Furnace Work.", Engineering, 24th March, 1922.
² Fuel Research Board, Technical Paper No. 7 : " Preliminary Experiments in the Low Temperature Carbonisation of Coal in Vertical Retorts." (H.M. Stationery Office.)

of the former to 40 per cent of the latter, this mixture having proved to give a satisfactory coke on carbonisation at 600° C. in the horizontal retorts at H.M. Fuel
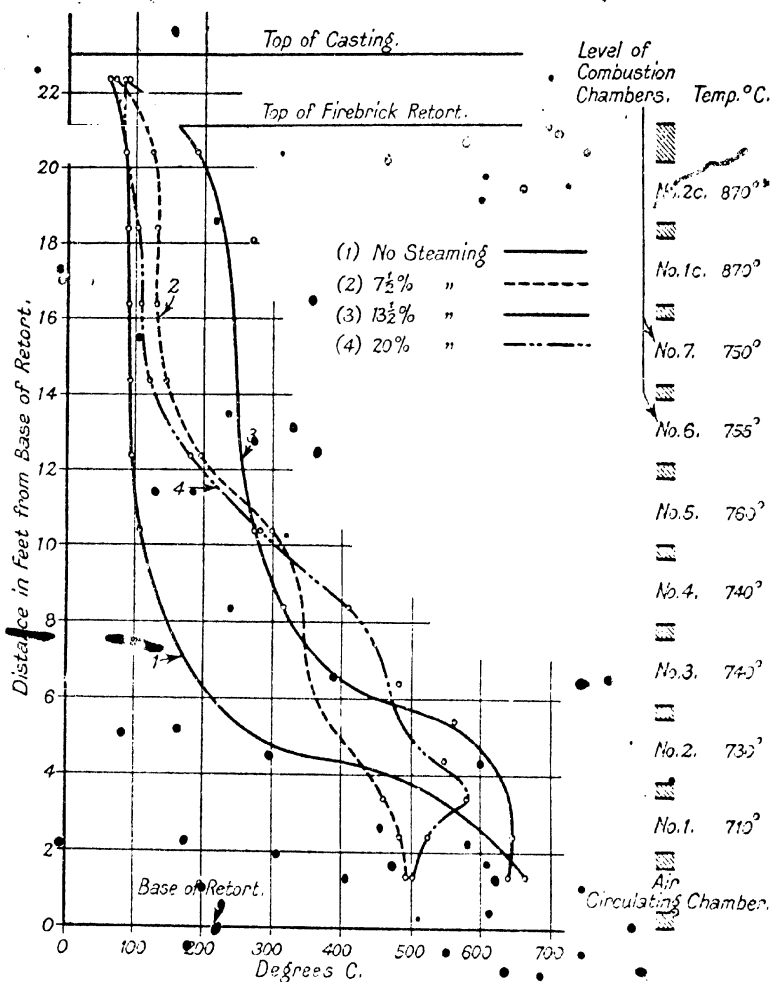
FIG. 14.—DISTRIBUTION OF TEMPERATURE IN No. 2 VERTICAL RETORT DURING LOW TEMPERATURE CARBONISING.

Research Station. A thermocouple was inserted in a long metal tube extending down the centre of the charge in the retort, and readings were taken at points 2 feet apart. The graphical representation of the results. given in Fig. 14, are

H

reproduced from the Report by permission of H.M. Stationery Office. The following conclusions from the graphs are also given in the Report :—

"With no steaming the temperature at the centre of the retort rises very slowly, while the coal traverses a distance of 12 feet. Thereafter it rises more rapidly for 5 feet, and then very rapidly at a distance of 16 feet, or 3 to 5 feet from the base of the retort. At the base it finally almost reaches the temperature of the combustion chamber immediately opposite.

"With steaming the rise of temperature is more rapid, an accelerated rise being obtained at about 10 feet from the top. The maximum attained is not so high by about 100° C. as with no steam, but appears at a level fully 2 feet higher in the retort.

"The tendency for the coal to hang up in the retorts for short periods, and then travel rapidly down for a short distance, causes a good deal of uncertainty in such temperature measurements, and it is only by continued poking that even reasonable results are obtainable."

TREATMENT OF MATERIAL IN THIN LAYERS

The fact of the slow rate of heat transference through coal is reflected in one of the primary principles in the designing of externally heated retorts, viz. that the material must be treated in relatively thin layers. There is a certain amount of experimental justification for assuming that the time required for portions in the interior of a mass of coal to be heated to a temperature within a few degrees of that of the heating surface varies roughly as the square of the distance from that surface of the portion considered. If therefore the flue temperature is kept constant, the time required to carbonise a mass of coal 12 inches thick is four times that required to carbonise a mass 6 inches thick, and even then the degree of carbonisation will not be uniform.

The bearing of this on the economics of the process lies in the fact that the throughput of a retort varies directly as the mass of charge, and inversely as the time of carbonisation. The throughput of a retort is a very important commercial consideration, and should be a maximum, so long as the products of carbonisation do not suffer thereby. In order to minimise the time of carbonisation there is therefore an upper limit of possibly 6 inches, to the desirable thickness of the charge within a retort, if the heating is to be effected solely by conduction from the outer surfaces, and if a commercial throughput is to be maintained. Many devices have been used, such as the subdivided trays of the Fuel Research Board and, as the webs in the Tozer retort, whereby the mass of material within the retort is broken by metal, which allows an increase in the cross-section of the retort without increasing the

thickness of the mass to be carbonised. These devices will be described in a later section.

The time for the complete carbonisation of coal depends, however, upon another equally important factor, viz. the time required to remove the gases and oil vapours from the charge. It may be surmised, for example, that the time of carbonisation of non-fusible coal is less than that of fusible coal. With non-fusible material the evolved vapours may escape freely through the interstitial spaces—unless indeed the charge is very closely packed—but, with fusible materials, the cementation period delays very considerably the time of carbonisation; since the evolved gases have to force their way through a viscous envelope. In the case of both fusible and non-fusible coals, the ratio of mass to surface exposed to heat, has an important bearing upon the time required for carbonisation.

The " time-element " for the removal of vapours must therefore be taken into consideration in determining the thickness of the layers for carbonisation. From the standpoint of the conduction of heat, it is advantageous for the thickness to be reduced ; from the standpoint of the time required for the completion of the chemical reactions in the coal, there is no advantage in using very thin layers.

EFFECT OF RADIANT HEAT

This position is strengthened when the effect of radiant heat is also taken into consideration. It has already been pointed out that under radiant heat carbonisation is effected so quickly that the surface particles have not time to fuse, and the resulting coke consists of completely detached particles. This is not a defect when the resulting fuel is to be briquetted, or can be used in powder form; but it prohibits the use of the fuel for domestic purposes when the proportion of breeze must of necessity be small.

EFFECTS OF STIRRING

There is a further possible method by which the time of carbonising may be reduced, viz. by continuously stirring or moving the material and bringing fresh surfaces into intimate contact with the hot sides of the retort, or with the heating gases. In the case of fusible and semi-fusible coals, especially if the motion occurs during the cementation period when the material is in a semi-plastic condition, this subjects the product to a certain amount of shearing action, and causes all the lumps of coke to disintegrate. The resulting residue is too fine to be serviceable ; or if not too fine, it is too friable for economic purposes. This is a fatal defect in any system which depends for its success on the sale of the residual coke as domestic fuel, for, owing to the high price of the binding material and the absence as yet of a satisfactory smokeless binder, the possibility of post-briquetting

must, in general, be seriously discounted from a commercial standpoint. Although small fuel is serviceable in a producer, there is a limit to the fineness when thus employed, on account of the increasing resistance offered by the fuel bed to the passage of air and steam as the proportion of " fines " becomes greater.

A further notable disadvantage of stirring is that the coke dust mixes with the oil products, making refining expensive and increasing the working difficulties through the blocking of the eduction pipes.

It is interesting in this connexion to note that, in the primary retort of the carbocoal process, the material is continually stirred and moved through the retort by the revolving paddles. A friable residue is the result, but as the material is briquetted with pitch before it is passed through the secondary retort, this technical disadvantage is nullified.

Reference may also be made to the work of Winmill[1] on the low temperature distillation of inferior coals. The coals tested had an ash content of more than 50 per cent. The apparatus used was designed for rapid carbonisation, without consideration of the effect upon the residuum. The material was therefore continuously and vigorously stirred. This certainly reduced the time of carbonisation, but also had the effect of reducing the residue to a fine powder. It was found, in consequence, that the choking of the eduction pipe of the retort was so troublesome that the speed of the stirrer had to be very appreciably reduced for optimum results.

EFFECTS OF INCREASING FLUE TEMPERATURE

The time of carbonisation of the charge may likewise be reduced by increasing the temperature of the flue gases. Clearly the rise in temperature of the centre of a charge will increase with the rise in temperature of the inner surface of the retort wall. Any increase of the flue temperature over a certain limit will, however, have very serious consequences. Either the outer layers become overheated, or the inner core is not sufficiently heated for correct carbonisation to occur. In either case there is a grave lack of uniformity in the coke residue at any cross-section, and the efficacy of the process is considerably diminished.

B. INTERNAL HEATING

CHARACTERISTICS OF HEAT-SUPPLYING MEDIA

When the heating is effected internally, the heating medium is passed into the retort, and, after giving up its heat to the charge, usually assists in carrying off the vapours of distillation. When the retort is externally heated, only the vapours of distillation need be condensed in order to recover the oil products ; but, when

---

[1] Winmill, T. F. , "The Low Temperature Distillation of Inferior Coal." *Journ. Soc. Chem. Ind.*, 1917, **36**, 912.

the retort is internally heated, the heating medium must also in general be passed through the condensing plant. It is desirable, therefore, that the volume of heating medium should be a minimum.

The medium supplying heat to the charge should possess the following two characteristics :—

(1) It should have a high specific heat, so that the requisite quantity of heat may be supplied with a minimum quantity of heating medium.

(2) It should be inert, so that combustion of the charge does not occur during carbonisation.

THE USE OF LIQUIDS OR SOLIDS AS A HEATING MEDIUM

Before dismissing the possibility of supplying heat to the charge by means of liquids, reference may be made to the process recently advocated by Mr. J. Stanley Morgan[1] in a paper read before the Northern Section of the Coke Oven Managers Association. In this process the powdered charge is immersed in a bath of molten lead, which is kept in motion in order to facilitate the heat transfer. The time factor of carbonisation is then very small, being of the order of two minutes as against hours by other processes. The whole of the charge is thus uniformly treated, and the gaseous vapours may be quickly removed. The process is, however, likely to present mechanical difficulties and is still in the experimental stage. The Caracristi retort, which also utilises molten lead as a heating medium, is described on page 209, but in this case the material is not, strictly speaking, heated internally.

The use of a solid material, such as heated sand, has also been suggested. Where oil and gas production only are required, as in the low temperature distillation of certain shales, the process is not without possibilities ; but in the case of coal, where the solid residue is one of the most important assets of the process, the difficulties of separating the sand after distillation precludes the possibility of such a method proving of commercial utility.

THE USE OF STEAM AS A HEATING MEDIUM

Steam has a high specific heat, but its use as a heating medium without extraneous assistance does not at present appear to be very promising, for three main reasons :—

(1) The practical difficulties in the supply of steam superheated to the degree necessary for the carbonisation of the charge.

(2) The necessity for provision of cooling plant of sufficient size to absorb the latent heat of the steam, as well as the sensible heat carried over in the

---

[1] Morgan, J. Stanley. "The Lead Bath Process of Low Temperature Carbonisation." *Colliery Guardian*, 13th April, 1923.

mixture of steam and carbonisation gases. Large and costly condensers would therefore be necessary.

(3) The difficulty that, unless the steam could be obtained as waste from some industrial undertaking, the latent heat would have to be supplied in addition to the heat necessary for carbonisation.

THE USE OF PRODUCER GAS AS A HEATING MEDIUM

A more satisfactory heating medium is stripped coal gas or producer gas. These gases are inert, or at least contain very slight traces of oxygen, and, after passing through the condenser, do not have any deleterious effects upon the volatile products taken from the gases after the distillation of the coal. They do, of course, reduce the calorific value of the resultant gases obtained from the retort. For economic reasons, producer gas is the medium generally employed in practice. There is in general a certain amount of dust carried by the producer gas, and its effect, which is perhaps not altogether a deleterious one, upon the carbonising charge must not be overlooked.

DIFFICULTIES IN INTERNAL HEATING

The main difficulty in internal heating is to secure uniform carbonisation of the charge. In the case of horizontal retorts, this can only be effected by stirring the charge continuously, or by other methods, some of which introduce grave practical difficulties in a plant of a commercial size. The effects of stirring the charge during carbonisation have already been discussed on page 115 and so need not be further referred to. They are sufficiently objectionable, however, to demand careful thought before stirrers are used with internal heating.

Rotary retorts of the "Sensible Heat" (Nielsen) type (page 251) are, however, in a different category. In this case the material is in continuous motion as it progresses through the retorts, but it is impelled by the force of gravity and not directly by mechanical means. There is therefore no tendency for the particles to disintegrate other than that experienced in any continuous plant. In this particular type of retort it is found in practice that the proportion of "fines" in the coke is not great.

In the case of a vertical type of retort the inert heating gas may readily be forced or drawn through the charge. It is difficult, however, to diffuse the gas adequately. It tends to pass up fissures through the material, and, taking the line of least resistance, fails to carbonise the material uniformly. The original Del Monte patent was based on this principle, which was indeed proposed by Parker as far back as 1890 ; and, although extremely satisfactory results could occasionally be obtained, the lack of uniformity in the residual products was so

great that this type of retort was eventually scrapped and an externally heated type adopted. It is doubtful, however, whether the experimental work in these early days was sufficiently under scientific supervision to form a criterion for future developments, since the later work of Maclaurin on a somewhat similar retort holds out great promise. Probably this difficulty may be overcome to a great extent by paying sufficient attention to the adequate sizing of the material.

Parr and Olin[1] also give interesting results obtained by this type of heating; but, since small-scale plant only was employed by them, the results are not strictly comparable with those obtained in a plant on a commercial scale.

AMOUNT OF GAS REQUIRED FOR INTERNAL HEATING

Owing to the small specific heat of producer gas it is necessary to determine very carefully whether the amount of gas necessary to carbonise the charge, to remove the products at their final temperatures and to provide for radiation losses can economically be obtained. We have already seen that in an internally heated system the heating gases dilute the gases of carbonisation, and it is therefore obvious that if the quantity of heating gas required is very great a point may be reached where the calorific value is so reduced as to make the mixed gases of no commercial value.

The work of both Maclaurin and Nielsen has shown that if proper steps be taken to reduce radiation and other losses to a minimum, a gas of commercial value may be obtained.

The question may best be dealt with by a consideration of the heat balance. If we take for example the case of a retort of the Nielsen type where the heating medium consists of hot producer gas made in a separate producer, we have entering the retort, the potential heat of the coal charged and the heat, potential, sensible and latent of the producer gas used. On the other hand, the heat leaving the retort comprises the potential heat in the products after cooling to the datum temperature, the heat both sensible and latent in the products as they leave the retort and which will be abstracted in the cooling arrangements, and the heat lost by radiation, etc. In order to obtain a gas of the highest possible calorific value it is essential, therefore, that the two latter terms should be reduced as far as is possible consistently with practical working. From the data indicated above, it is possible to estimate the volume of producer gas at any given temperature required per ton of coal carbonised, and so to calculate with sufficient accuracy for the purpose of a preliminary investigation the degree of dilution to which the rich gas of carbonisation will be subjected.

[1] Parr and Olin. "The Coking of Coal at Low Temperature." *Bulletin* No. 60, University of Illinois, 1912.

COMPARISON OF EXTERNAL AND INTERNAL SYSTEMS
OF HEATING

Having outlined the difficulties of each of these systems of heating, their relative advantages and disadvantages may now be summarised.

Theoretically, internal heating is more advantageous than external heating, since the heating medium is brought into direct contact with the material to be carbonised, and the heat transmission is very effective and rapid. It is not necessary to treat material in thin layers, which, as already pointed out, is a desirable feature in retorts heated externally. The simple retort thus rendered possible results in a reduction in the capital cost of plant and in maintenance and labour charges. A further advantage is to be found in the fact that the passage of the heating medium through the retort usually assists in carrying off the vapours formed.

Among the disadvantages of internal heating may be mentioned :—

(1) The additional condensing plant required through the mixing of the heating medium with the gaseous products of distillation.

(2) (a) The possibility of the material not being coked uniformly in a vertical retort ; (b) the necessity of employing a long horizontal retort in order to abstract the heat from the heating medium.

(3) The difficulty of controlling the temperature gradient through the retort if producer gas is used as the heating medium.

(4) The rich gas ordinarily obtained by low temperature carbonisation is mixed with the poorer producer gas used as a heating medium, and the calorific value of the mixture, though higher than that of producer gas, is considerably less than that obtained from an externally heated retort. The extra quantity of gas may not compensate commercially for its loss of calorific value, and it usually requires a special market.

The main advantage of heating retorts externally is that this method has been very extensively employed in ordinary gasworks and shale oil practice, and that the published data of the experience gained can be used as a guide in solving some of the difficult problems of low temperature carbonisation. The plant can also be designed so that the process is under more direct control than in the case of an internally heated plant.

TECHNICAL PROBLEMS—THE EFFECTS OF STEAM AND GASEOUS PRESSURE ON COALS CARBONISED AT LOW TEMPERATURES

A. THE EFFECTS OF STEAM

ACTIONS AND REACTIONS OF STEAM AND HEATED CARBON

It is well known that steam will react when passed over certain strongly heated reducing agents. The oxygen in the steam combines with the reducing agent, the final products of the reaction being formed in definite relative proportions which depend upon the conditions, such as temperature and concentration, or percentage volume, of the various gases present, and also upon whether there is sufficient time for equilibrium to be established. When the reducing agent is carbon, the products of the reaction are hydrogen, carbon monoxide and carbon dioxide, which will be mixed with a certain proportion of the steam left undecomposed. Two primary reactions take place, according to the temperature of the carbon and the concentration of the gases, which may be represented by the equations :

$$H_2O + C \rightleftharpoons H_2 + CO \qquad . \qquad . \qquad . \qquad (1)$$
$$2H_2O + C \rightleftharpoons 2H_2 + CO_2 \qquad . \qquad . \qquad . \qquad (2)$$
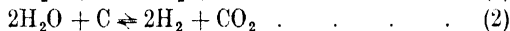
In addition to the above primary reactions, there are many others taking place between the various products and the carbon and steam, of which perhaps the most important are :—

$$C + CO_2 \rightleftharpoons 2CO \qquad . \qquad . \qquad . \qquad (3)$$
$$H_2O + CO \rightleftharpoons H_2 + CO_2 \qquad . \qquad . \qquad . \qquad (4)$$
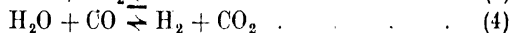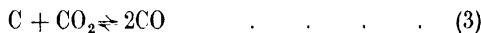
As indicated by the equations, each of the above four reactions is a balanced reaction, i.e. one capable of proceeding in either of two directions according to the existing physical conditions.

If any of the above reactions took place alone, using definite amounts of the reacting materials, the final mixture would adjust itself at any definite temperature according to the law of mass action, until the reacting materials and products were present in certain definite proportions, provided sufficient time were given to allow equilibrium to be established.

In the case of the reaction shown in equation (4), which is known as the water gas reaction, if we start with definite amounts of the four reacting gases, equilibrium will be established when the product of the percentages by volume of carbon monoxide and water is equal to the product of the percentages by volume of carbon dioxide and hydrogen, multiplied by a constant K.

$$\text{or } K = \frac{\text{Percentage of CO} \times \text{percentage of } H_2O}{\text{Percentage of } CO_2 \times \text{percentage of } H_2}$$

The numerical value of K, the equilibrium constant, is a fixed quantity for each temperature.[1]

Experimental values of K for given temperatures are as follows :—

Temperature °C.	700	800	900	1000	1100	1200
K	0·55	0·86	1·25	1·65	2·10	2·56

In any process involving a number of reactions, taking place simultaneously, the final product will obviously depend largely upon the relative speed at which the equilibrium of the separate reactions tends to establish itself. In the water gas process the temperature coefficient of reaction (1) is greater than that of reaction (2) with the result that production by reaction (1) is favoured at high temperatures. It can also be shown that at temperatures above 600° C. the equilibrium of equation (4) is very rapidly established. At temperatures above 1000° C. therefore, we find that the effects of (1) and (4) predominate, and those of (2) and (3) are comparatively small. This also is the case at the lower temperatures if pure carbon is used, but if coke is employed reaction (2) is greatly accelerated, probably by the presence of catalytic materials, such as iron oxides in the coke. The velocity of reaction (3) is comparatively slow, and its effects are negligible.

In water gas practice, therefore, reactions (1) and (4) appear to predominate above 1000° C. whilst reactions (2) and (4) predominate at temperatures below, say, 700° C.

An examination of (1) and (4), together with the given values of K, show that the percentage of carbon monoxide is high and that of carbon dioxide low at high temperatures, whilst the percentage of carbon dioxide is high and that of carbon monoxide low at low temperatures (700° C.).

It will also be seen that, if a water gas rich in carbon monoxide is required, an excess of steam is to be avoided, since by equation (4) the steam when in equilibrium is in inverse ratio to the carbon monoxide ; also excess steam, by lowering the temperature, reduces the value of K and so again tends to the formation of excess carbon dioxide.

PRODUCTION OF WATER GAS AND POWER GAS

In practice water gas is formed by passing the steam through a deep incandescent fuel bed in a producer. A certain amount of carbon dioxide is formed in

[1] The relation between K, the heat of reaction and the temperature is defined by the Van 'tHoff equation $\dfrac{d \log_e K}{d T} = \dfrac{Q}{R \cdot T^2}$ in which Q is the (positive) heat of reaction at the absolute temperature T, and R is the gas constant ($=1.985$ calories). For further details the reader is referred to any standard book on physical chemistry.

the lower part of the bed, in addition to the carbon monoxide represented by equation (2), according to the equation :—

$$2H_2O + C = CO_2 + 2H_2 \quad . \quad . \quad . \quad . \quad (5)$$
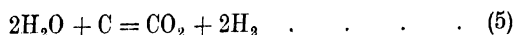
This carbon dioxide is subsequently reduced to carbon monoxide while passing through the upper part of the fuel bed, according to the equation :—

$$CO_2 + C = 2CO \quad . \quad . \quad . \quad . \quad (6)$$

Apart therefore from the impurities in the water gas due to the fact that the fuel used may only contain 90 per cent or less carbon, it is to be expected that circumstances will arise in practice when the whole of the carbon dioxide is not reduced to carbon monoxide. There is always therefore a certain amount of carbon dioxide and other inerts in the resulting gases.

The other factors to be taken into consideration are :

(a) The reactions (1), (2) and (3) are endothermic in character, and

(b) They do not occur if the temperature of the carbon falls below a certain minimum. In the case of reaction (2), that is in the formation of carbon dioxide, the minimum temperature is about 600° C. In the formation of carbon monoxide, a slightly higher temperature is necessary. Unless therefore heat is supplied to keep the temperature of the fuel above the minimum given, these reactions do not occur.

Water gas producers are generally worked upon an intermittent system, that is, the gas making period, termed the "run" during which the temperature of the fuel falls considerably. is followed by a period of heat recuperation, technically known as the "blow." During the "blow," the steam is replaced by a blast of air, the oxygen of which combines with the carbon, mainly to form carbon dioxide. This is an exothermic reaction, and by the continuance of the "blow" for a certain period the temperature of the fuel and the surrounding brickwork is increased preparatory to the gas-making period again commencing.

Alternatively a power gas, though not water gas, may be produced by passing steam and air (or oxygen) continuously through a producer. When correct proportions of steam and air are admitted, the endothermicity of one set of reactions may be balanced by the exothermicity of the other, and the temperature of the fuel thus kept constant. While it is not necessary to discuss further the production of water gas or of power gas in producers, the above digression is perhaps permissible in leading to a clear comprehension of the effects on the charge in low temperature carbonisation when, as is sometimes done, steam in limited quantities is fed to the discharge mechanism in order to keep the metal parts cool and to prevent the escape of gas during the period when the retort doors are open.

ADVANTAGES OF STEAMING FOR HIGH TEMPERATURE CARBONISATION

The advantages of steaming for high temperature carbonisation are unquestionable. The work of the Fuel Research Board[1] and that of the Gas Investigation Committee of the Institution of Gas Engineers,[2] show that increased therms in the form of gas, and increased quantities of tar and ammoniacal liquor are to be obtained by this method, although the quantity of coke is reduced. Sir George Beilby, from his long experience in steaming charges of shale in the Scottish shale oil industry, as well as from results obtained at H.M. Fuel Research Station, is of the opinion that high temperature vertical retorts should never be worked with less than 5 per cent of steam, and this percentage has in fact been adopted as the datum line for all future experiments in this connexion at H.M. Fuel Research Station.

One factor of great interest in the results obtained at H.M. Fuel Research Station which should be mentioned at this stage is that, in all the coals tested, an increased yield of therms in the gas was obtained at a decreased expenditure of heat, with the use of 3 per cent of steam as compared with the yield obtained when no steam is supplied. The explanation lies in the fact that heat is abstracted from the hot coke at the bottom of the retort owing to the action of the steam which is converted into water gas. There is usually no necessity for quenching the coke, which leaves the retort at a much lower temperature when a small amount of steam is used than when there is no steaming.

EFFECTS OF STEAMING IN LOW TEMPERATURE PROCESSES

The advantages of steaming for low temperature carbonisation are, however, more problematical. If the temperature of the charge does not exceed 600° C. the decomposition of steam by carbon is practically negligible. In any case, the formation of carbon dioxide rather than carbon monoxide, is favoured, and thus the calorific value of the gas is reduced. Any beneficial action of the steam is probably either physical or mechanical rather than chemical. The continuous or occasional admission of steam near the coke exit of the retort cools the coke prior to its removal, and thus tends to increase the efficiency of the process. It also protects the material of the retort by reducing the temperature at the coke exit where otherwise the heat would often be greatest. A further advantage of steaming is that the heat energy absorbed by the steam in its earlier path through the retort is diffused at a later stage, and the steam may therefore be said to assist in conveying

[1] Fuel Research Board. Report for years 1920, 1921 ; 'First Section : "Steaming in Vertical Gas Retorts." (H.M. Stationery Office.)

[2] Institution of Gas Engineers : Fourth Report of the Gas Investigation Committee, 1920.

heat uniformly to the charge and in securing a more satisfactory heat gradient through the retort than would otherwise be obtained. Finally, the passage of steam through the charge assists in sweeping away the vapours of carbonisation as they are formed. If coal contains little moisture the ammonia escapes condensation and a scrubber is necessary. If steam is present the ammonia is more completely condensed without scrubbing. In using a scrubber the liquor would be too weak to be of any value.

When the carbonising temperature is in the neighbourhood of 700-750° C. chemical reactions do probably exert some influence on the resulting products of carbonisation, partly in a slight increase in the gas yield, and partly in a slight increase in the amount of ammonia recovered. The increase in the ammonia yield at high temperatures due to steaming is well known, and at lower temperatures there is also a slight advantage in steaming. The amount of ammonia yield in low temperature work is, however, so small even with steaming, that it is probably only in plants with a large throughput that steaming may be said to be advantageous in this respect.

It is also well known in the shale industry that steaming has the effect of preserving the primary products of distillation at low temperatures, and this factor is also of importance in low temperature carbonising processes.

ADVANTAGES AND DISADVANTAGES OF STEAMING

To summarise, the advantages of using a small amount of steam in low temperature work may be said to be :—.

(1) Steaming assists in distributing the heat uniformly through the charge.
(2) Steaming increases slightly the yield of gas, but only at the higher temperatures of carbonisation.
(3) Steaming facilitates the recovery of ammonia, also at the higher temperatures of carbonisation.
(4) Steaming assists in removing the gaseous products of distillation.
(5) Steaming increases slightly the efficiency of the process by cooling the coke before its withdrawal from the retort.

On the other hand, the disadvantages of steaming are : —

(1) The cost of the steam and the extra cost of maintenance and manipulation, i.e. power, extra fittings, supervision, etc.
(2) Complete loss of latent heat of steam in the condensing processes.

OTHER PRACTICAL CONSIDERATIONS

It should be pointed out that the design of retort has an important bearing on deciding whether steaming should or should not be adopted. Quite apart from the question whether or not steaming is beneficial, horizontal retorts do not in general allow the steam to be brought into that intimate contact with a mass of incandescent coke which is desirable for the full effects of steaming to be realised. In the case of the Chiswick retort for example (page 199), the introduction of steam for uniform reaction upon the whole of the coke residue is hardly feasible. Steaming may more effectively be employed in vertical retorts, but even then in particular cases the beneficial effects of steaming could not be fully realised. It appears therefore upon close examination, that local considerations will mainly affect the decision as to whether or not it is desirable to use a small quantity of steam in any particular plant.

B. THE EFFECTS OF GASEOUS PRESSURE

EFFECTS OF VERY LOW PRESSURES

The effects of the extreme limits of pressure, either very low or very high, on coal during carbonisation seem to be marked. The earliest record of the effects of very low pressures appears to be in the Patent Office records for 1905. In this year W. S. Simpson obtained a patent on the claim that sulphur and other impurities are removed from coke by creating a vacuum in the vessel containing the coke in a highly heated state. In 1906 he obtained another patent on the claim that, when carbonaceous materials are heated in a practically complete vacuum, a superior quality of charcoal is produced and impurities removed. Use has been made of low pressure in at least one process of low temperature carbonisation put forward, viz. the Tozer retort. (See page 197.)

Unfortunately few results of experimental investigation on the effects of very low pressures have been published; Taylor and Porter[1] have found that a bituminous coal which yielded a light friable coke when slowly heated at atmospheric pressure yielded a dense coke when heated at a pressure of less than 30 m.m. A probable explanation of this phenomenon is that the gas is rapidly removed when formed, and therefore there is not time for the development of a large bubble structure.

[1] Taylor and Porter. "The Primary Volatile Products of the Carbonisation of Coal." Tech. Paper 140, Amer. Bureau of Mines.

PRACTICAL ASPECTS OF A VACUUM SYSTEM OF CARBONISATION

There are, however, other advantages of a vacuum system which are less questionable. These may be thus briefly summarised :—

(1) The removal of the gaseous vapours is facilitated, and any heavy tars formed need not condense within the retort to the same extent as in other processes.

(2) The removal of air in the retort, particularly in an intermittent process, reduces the likelihood of the oxidation of the products of carbonisation.

(3) The reduction of the pressure within the retort may possibly increase the virtual temperature of carbonisation by the reduction of the boiling points of the volatile substances once their temperature of formation is reached. Hence the same results are possibly obtained by a vacuum process with a flue temperature lower than that of an atmospheric process, *ceteris paribus*.

It must be said, however, that the above advantages are likely to be theoretical rather than practical, and claims for increased gas or oil yields by vacuum processes have not been fully substantiated. On the other hand, the disadvantages of working on a commercial scale with a high vacuum are important. These may be thus summarised :—

(1) Greater capital cost of plant.

(2) Increased operating costs, due to the extra pumps, etc.

(3) Increased difficulties in maintaining air-tight joints, some of which must be frequently broken and re-made ; as, for example, at the inlet and outlet of the charge.

(4) Danger of air-leakage inwards, with the consequent dilution of the rich low temperature gas and possibly the attendant risk of accidental explosions.

It would appear therefore that the only substantial commercial advantage of a vacuum process consists in certain cases in the production of a dense coke and a tar which has suffered the minimum amount of cracking within the retort. As this advantage may be secured by other and perhaps simpler methods, it is not surprising that the development of a vacuum system for a commercial process of low temperature carbonisation appears to be a somewhat remote contingency.

EFFECTS OF HIGH PRESSURE

It has been known for some time that the distillation of petroleum under pressure serves to break down or "crack " the high-boiling fractions, and so may increase the value of the products. Nearly thirty years ago Dewar and Redwood patented a process based on this principle, though commercial use was not made of the patent. On account of the increasing demand for gasoline for the automobile

industry, and the desirability of obtaining substantial yields of low-boiling fractions in the distillation of petroleum, the problem was taken up by Burton in America about 1910. Burton's first experiments were in the direction of superheating the high-boiling petroleum residues at atmospheric pressures. It was found, however, that "cracking" by this method turned the aliphatic hydrocarbons into aromatic bodies and fixed gases ; that is, the general molecular structure of the original material was altered. The use of reagents and catalysers was fairly successful, but involved a substantial loss of oil in the process, and was, moreover, costly. Eventually a process of distillation under a pressure of about five atmospheres was evolved in the laboratory, which on being worked on a commercial scale gave good results, and the pressure-still industry is now firmly established.

There are, however, limits to the Burton process and its modifications. Cracking processes are not uniformly successful with heavy oils and with those containing high percentages of asphalt. In these cases there is a tendency for hydrogen to separate and to leave a deposit of carbon. The deposition of carbon is not only troublesome mechanically, but involves a serious reduction in the efficiency of the process.

The deposition of carbon may be reduced by the introduction of hydrogen into the process, and Bergius claims that, by a cracking process at high temperatures and high pressures in the presence of hydrogen, there is a reduction in the carbon deposit without having recourse to catalysts. The pressures used by Bergius are, however, so great as to preclude any immediate application of his system for the hydrogenation of oil and coal to the problems of low temperature carbonisation.

PRACTICAL ASPECTS OF A HIGH PRESSURE SYSTEM OF CARBONISATION

The most interesting development of the work of Bergius in connexion with the carbonisation of coal is in the liquefaction of coal by the method which has already been described (page 47). It is stated that the technical difficulties of the process have been largely overcome, and that a plant on a semi-works scale is now in operation at Mannheim for the conversion of bituminous coal into light oils. Granting that the work has been successful from the laboratory standpoint, and that suitable precautions can be taken to minimise the serious risks of explosions and fires, it would appear that the capital cost of the plant must necessarily be high, and the working costs, including the cost of the hydrogen, are likely to be excessive. This process, although of intense scientific interest, can therefore scarcely be of great industrial importance until it has been suitably developed to meet the economic conditions of the day.

PRESSURES ADOPTED IN PRACTICE

In practice the withdrawal of the gas and oil vapours is generally secured by keeping the retort under a pressure which is slightly below atmospheric. The extent to which it is desirable to reduce the pressure below the atmospheric pressure depends very largely upon the number and type of joints on the plant which must be kept air-tight, and also upon the material used for the retort. With the exception of the two processes to which reference has already been made, it is found that plants are usually arranged to work under a partial vacuum which does not greatly exceed 2 inches of water pressure.

CHAPTER X

OTHER TECHNICAL PROBLEMS

INTRODUCTORY

HAVING now dealt with some of the broad general principles which affect the design or selection of a plant for low temperature carbonisation, it is desirable to consider shortly some other features of the process. Many of these have already been referred to in the more general discussions. The various points for consideration in a subject such as the present are so interrelated as to make it difficult to take them singly without a certain amount of duplication and overlapping in the various sections. The advantages of this method of treatment are, however, very great, and the present chapter will be devoted to a sectionalised treatment of the following points, some of which have already appeared as side issues in the discussion of wider aspects of the problem, viz. :

(a) The effect of the change of volume of the charge.
(b) The withdrawal of the oil vapours.
(c) The control of the temperature of the charge.
(d) The conservation of heat.
(e) Simplicity of the construction and operation of the retort.
(f) The reduction of capital, maintenance and labour costs.

SWELLING AND NON-SWELLING COALS

Amongst other classifications we have seen that, for the purposes of low temperature carbonisation, coal may be divided into two groups according to the relative volumes of the original coal and the resulting coke, after carbonisation at low temperatures. In the first group may be placed the so-called " swelling " or " expanding " coals, the volume of the resulting coke being greater than that of the original coal. The second group consists of the non-swelling coals, in which the volume of the resulting coke is equal to or smaller than that of the original coal.

It should be noted that, although these groups are sometimes termed " expanding " and " non-expanding," these descriptions are somewhat unsatisfactory, as they may leave the wrong impression that the actual coke substance is of different densities in the two cases. The coke substances seem to have in fact equal densities, and the apparent " swelling " or " expansion " is due to differences of structure. It is not necessary, at this stage, to enlarge upon the point, and enter upon a full discussion of coke structure and the theories of coking, a study which will be better treated in a separate chapter. It may, however, be desirable in passing to refer to the microscopical work of Sir George Beilby, who showed that the cell structure of coke is due to the evolution of gas bubbles from the fused or partially fused coal substances. The first stage in the structure is the formation of foam, in which

each bubble is a self-contained cell. Eventually mutual perforation of the bubbles occurs at the points of contact, and through the holes thus formed the gases ultimately escape to the exterior of the sponge-like mass.

SWELLING COALS

When coking coals are heated, they contract a little at first, but swell on further heating. At a still higher temperature however, they undergo a secondary contraction which is characteristic of coking proper. The initial contraction is possibly due to the melting of certain material which occurs at a temperature of 350° C.–450° C. according to the constituents in the coal. If the temperature of the coal is uniform throughout the mass, the whole charge is fused together. When more heat is applied gas is given off, forming bubbles which increase the volume of the substance by an amount greater than the original contraction. As the temperature is further increased and the volatile products are driven off, the secondary contraction occurs.

It is well known, however, that the swelling of coals at the lower temperatures of carbonisation causes difficulties in the withdrawal of the coke made at those temperatures. When the swelling is restricted, a dense coke suitable for use as a domestic fuel is the outcome. The swelling may, however, be so much greater than the initial contraction that a considerable pressure is exerted by the coke on the walls of the retort. The charge may thus be held up at the time of discharge. This difficulty has been experienced from the earliest times in the history of low temperature carbonisation, and constitutes a factor which must be recognised in the design of any type of retort or in the development of any particular process. It may be overcome by mechanical means, as, for example, in the collapsible moulds of Richards and Pringle,[1] or in the collapsible centre plates of the Low Temperature Carbonisation Company's retort at Barnsley. The fact that these devices were adopted after experiments had been conducted on simpler types of retorting arrangements is in itself a sufficient indication that the problem demands attention in any full-scale plant. The more recent work on this question of obtaining a dense coke indicates, however, that a solution may be obtained in a simpler manner by physical or chemical rather than by mechanical means. In considering any mechanical device for reducing the difficulties due to the swelling of the charge, it should be borne in mind that labour costs must be reduced to the minimum.

NON-SWELLING COALS

In the case of the non-swelling coals and such carbonaceous materials as shale, cannel, etc., the coke residue is usually non-coherent and friable, and the problem

[1] Richards and Pringle. Pat. Spec. 27828 (1909).

of the carbonisation of the material is considerably simplified. It may be stated as axiomatic, indeed, that a plant which does not function for non-swelling coals will be useless for swelling coals. The special problems of the non-swelling coals need not therefore be discussed in this section.

PRELIMINARY TREATMENT OF SWELLING COALS

It has already been indicated that a solution of one of the difficulties in the carbonisation of swelling coal lies in the stopping of the swelling by other than mechanical means. Three methods, involving a preliminary treatment of the coal, have been suggested, viz. :—

(1) Blending caking with non-caking coal.
(2) Blending coal with coke breeze.
(3) A preliminary heat treatment of the coal.

Aspects of these will be dealt with in the succeeding paragraphs, but a fuller examination will be made in Chapter XI.

THE BLENDING OF COAL

It has been found that the blending of coal, i.e. the intimate mixing of fusible and less fusible coal, is a ready method of checking excessive foaming during carbonisation, and of ensuring that the resulting coke occupies a smaller volume than the original coal. By arranging for the shrinkage of coal instead of relying upon the confinement of coal in a restricted space during carbonisation, one of the most serious difficulties in the carbonisation of fusible coals is removed. It is interesting to note, however, that the proposal of blending coal is of by no means recent origin. The idea seems to have been first covered by Cruickshanks in a patent granted in 1841.

At an early stage in the work at East Greenwich the Fuel Research Board utilised the method of blending swelling and non-swelling coals in order to produce a dense coke which could be discharged with facility.

Roberts[1] obviated the swelling of coal by utilising the excess binder to bind inert material, say coke breeze. The presence of any appreciable quantity of coke breeze, however, reduces the amount of virgin coal carbonised per charge, and therefore reduces also the real throughput of the retort.

In the case of the Barnsley plant, the charge admitted to the retort is a mixture of coking and non-coking coals in the proportion of about 30 : 70. The use of non-coking coal has not the disadvantage of coke mentioned above, though the

[1] Roberts, J. "The Carbonisation of Coal at Low Temperature." *Proc. N. of Eng. Inst. of Min. and Mech. Eng.*, Aug. 6th, 1921.

expense of obtaining the necessary supplies of coal for blending, and the further trouble and expense of adequately mixing the coals, have to be faced when considering the commercial possibilities of the method.

PRESENT POSITION REGARDING BLENDING

The following quotation from the previously mentioned Report[1] may be given as representing the position at the present time regarding blending :—

"We have already referred to the fundamental importance of securing the volatile products, gas and oil, in maximum quantity and of the best quality if low temperature carbonisation is ever to be carried out on a really large industrial scale. This need not be taken as implying that the mixture of coke with coal before carbonisation is in no case desirable, but only that the use of this method must be carefully scrutinised before it is adopted in any particular case in which its economic advantages may possibly outweigh its disadvantages. That there are cases of this kind no expert in carbonisation could deny.

"We are well aware of the practical and economic difficulties which are involved in any general adoption of ' coal blending ' in place of the present crude methods of using so heterogeneous a material as coal as when it is mined. We believe, however, that it will be increasingly recognised that the advantages of insuring homogeneity and uniformity in any form of solid fuel (thereby raising the availability of its potential heat value to a much higher degree) are so great that these difficulties will ultimately be faced and overcome."

THE PREVENTION OF EXPANSION BY OXIDATION OR BY PRELIMINARY HEAT TREATMENT

Several other methods have been suggested from time to time for modifying the properties of swelling coals by preliminary treatment. It is well known, for instance, that, if certain types of coal are subjected to oxidation, their coking power may be considerably modified, and indeed completely destroyed. In a discussion on the spontaneous combustion of coal, Bone[2] describes some experiments on the absorption of oxygen by a Durham coking coal, and a Barnsley " hard steam " coal of almost the same ultimate composition. In these experiments oxygen dried over calcium chloride was circulated continuously over a weighed sample of ground coal previously dried in vacuo over sulphuric acid. The experiments continued over three to four weeks at a mean temperature of 108° C. In both cases the two oxides of carbon were continuously produced and oxygen was absorbed, although action was

---

[1] Fuel Research Board. Report for Years 1920, 1921; Second Section: "Low Temperature Carbonisation." (H.M. Stationery Office.)

[2] Bone, W. A. : Coal and its Scientific Uses. Chap. IX. (Longmans, Green & Co.)

more marked in the case of the Barnsley coal. Bone points out that the "coking properties of each coal were completely destroyed by the treatment to which it had been subjected."

Roberts[1] describes experiments on Silkstone coal ground to 30 mesh and heated at temperatures up to 110° C. for a considerable period in an air oven, in which he was able to produce a material which on subsequent coking under low temperature conditions yielded a good low temperature coke, although the original coal was of the type which could be classed as strongly swelling.

As far as can be seen at present, however, the method of oxidation does not seem likely to be of commercial application, owing to the time required for the preliminary treatment and the difficulty likely to be experienced in controlling the amount of oxidation. Again, the method is only applicable to coals which possess an excess of binding constituents. The quantity of such coals available is limited, and from the broadest point of view it would appear more desirable to utilise the excess binder to produce good quality coke from materials which, while yielding considerable quantities of oils and gas, are deficient in binder, and so do not by themselves produce smokeless fuel.

Reference has already been made in Chapter V to the work of Dr. Illingworth, who has succeeded, by means of a small scale plant, in producing hard low tempera-ture coke by submitting coals otherwise unsuitable for low temperature carbonisa-tion in their raw state to a preliminary heat treatment. Illingworth,[2] adopting the Seyler classification, states that coke structure is formed in stages by reason of the successive decomposition of the several types of "gamma compound," or cementing ingredient, and that the carbonaceous coals are one-stage coals. The meta-bitu-minous coals are also one-stage coals, but tend to become two-stage coals as the C/H ratio decreases. The ortho-bituminous coals are definitely two-stage coals, tending to become three-stage coals with decrease in C/H ratio, while the para-bituminous and lignitous coals are three- and four-stage coals respectively.

Illingworth also points out that, where a low gradient of temperature is employed, as in the subject under consideration, the stages in the process are well marked. If the mass be very plastic over the period of the earlier stages, the products evolved during these stages will pass away freely, and so will have little effect in causing the charge to swell. If the mass is very viscous, these products are trapped and tend to cause swelling.

Illingworth states that "plasticity is determined by the amount of gamma compound present," and terms the temperature at which the "gamma content" is finally decomposed "the critical temperature." If the critical temperature is

[1] Roberts, J.: "Economic Aspects of Low Temperature Carbonisation." *Proc. S. Wales Inst. Eng.*, 1922.
[2] Illingworth, S. R.: "Low Temperature Carbonisation of Coal." *Proc. S. Wales Inst. Eng.*, 1922.
Illingworth, S. R.: "Researches on the Constitution of Coal." *The Colliery Guardian* Co. Ltd.

low, and the " gamma content " is also low, the swelling of the coal is small, and such coals give good low temperature coke.

In the cases of two- or three-stage coals which contain binding material which decomposes over a wide range of temperature, Illingworth eliminates the less stable constituents by suitable heat treatment, so that the more stable only are operative in the subsequent carbonisation. Such coals are thereby converted to one-stage coals, with the result that they then produce robust types of low temperature coke. Illingworth has also found that the less stable constituents have very inferior powers of agglutination, and that in eliminating such from the coal the coking qualities are not seriously impaired. He also considers that the preheating of coal under suitable conditions produces a much better fuel in all cases, even from blends of coking and non-coking coal.

The method of preparation of a single coal by pre-heat treatment of course shares the disadvantage of processes which destroy some of the binding material in an attempt to obviate excessive foaming, but offers an alternative method of preparation in cases where a supply of non-coking coal is not readily obtainable.

THE WITHDRAWAL OF THE OIL VAPOURS

The early withdrawal of the oil vapours from the retort after their formation is desirable for both physical and chemical reasons. From the physical standpoint the early withdrawal of the hydrocarbon gases means the saving of a certain amount of heat. Any heat supplied over the minimum necessary to form the vapours and gases is wasted, since the gases are in general condensed and cooled before being further utilised. A low final temperature of the evolved gases therefore not only throws less work on the condensing plant, but also assists in increasing the efficiency of the heating arrangements and in decreasing the time of carbonisation.

From the chemical standpoint the gases and vapours should not be subjected for long periods to radiant heat, and it is desirable to prevent them from coming into contact with the hot walls of the retort, or from passing for any appreciable distance through the hot charge, in order to avoid undue " cracking " and decomposition of the gases and oil vapours. It is well known that the volatile products of one temperature are readily decomposed at a higher temperature. This may be sometimes desirable in gas-works practice, when gas of the declared calorific value is the main product, and is sometimes indeed, deliberately done in order to " crack " the higher hydrocarbons into the lower with the deposition also of free carbon. In the case of low temperature carbonisation, it is necessary to reduce the free space above the charge to a minimum in order that the oil vapours will be withdrawn from the retort as speedily and uniformly as possible after their formation.

Oil vapours can in general, be withdrawn from stationary horizontal retorts sooner after formation than from rotating and vertical retorts. If the material is progressively advanced through a horizontal non-rotating retort, eduction pipes can be conveniently placed for the withdrawal of gases. It is true that a fractional distillation occurs in a continuous vertical high temperature retort since, with an open charge, the products of carbonisation have to pass through no higher temperature than that at which they are evolved. It is, however, important to differentiate between the behaviour of the condensible and the non-condensible gases during this process. The non-condensible gases are probably not affected, but the condensible gases, passing upwards through the cool portion of the retort, will be subjected to a complex series of condensations and re-evaporations which cannot be beneficial to the lighter fractions of the oil vapours. On the other hand, by having the eduction pipes suitably located, the condensible and non-condensible vapours can, in the case of a horizontal retort, be withdrawn into a condensing chamber at practically the same temperature at which they are formed, and with a minimum of condensation within the retort. It is quite certain that the fractional distillation of the continuous vertical high temperature retort does not in practice give the same quantity or quality of oil yield which may be obtained by low temperature carbonisation.

Although eduction pipes can be more suitably placed on horizontal than on vertical retorts, the fractional condensation of the oil vapours should not be attempted. It has been demonstrated again and again that it is not possible to condense the oil vapours fractionally with any degree of commercial success.

THE CONTROL OF THE TEMPERATURE OF THE CHARGE

Sufficient has already been said about the carbonisation of coal to indicate the desirability in low temperature work of keeping the temperature of the charge below an upper limit. It is as desirable also to prevent local overheating, or indeed local underheating. The use of a very high flue temperature in an externally heated retort in order to reduce the time of carbonisation of the charge would demand a very careful control over the process which could probably not be realised in practice.

It would appear that temperature control is likely to be exercised better in a continuous than in an intermittent process. In a continuous process not only should the temperature of the heating medium be under control, but also its volume; or in other words, arrangements should be made so that the quantity of heat available for carbonising purposes can be adjusted from time to time to allow for variations in the quantity and physical characteristics of the throughput. The compactness of a retort is a further feature which facilitates a good control over the temperature.

No attempts have yet been made on a full-scale plant to vary the heat supply to a charge during different phases of its carbonisation. The effects of slow and of rapid heating of coal at various temperatures have already been discussed (page 96), but so far the beneficial effects of slow heating up to 200° C. and of rapid heating after 400° C. have not been utilised on any practical scale. Undoubtedly there are difficulties in carrying out a suggestion of this sort, but these difficulties do not appear to be insuperable in certain designs of retort which have already been suggested.

It should be pointed out that variations in the heating temperature are less likely to occur in fire-brick than in metal retorts because of the quantity of heat stored in the larger bulk of the former type of retort. This advantage is more than offset, however, by the greater liability of the brick retort to develop leaks.

THE CONSERVATION OF HEAT

Heating losses by radiation and convection, and in the flue gases and the discharged coke should be reduced to a minimum. If it is absolutely essential to utilise the non-condensible gases to heat the retort, particular attention should be paid to the location and design of the burners. In many of the processes for low temperature carbonisation which have been tried, particularly in retorts of the horizontal type, this seems to be a very common source of weakness.

From the standpoint of heat conservation it would appear that a continuous process is likely to be more successful than one of an intermittent nature. In the latter the retort is cooled each time a fresh charge is put in, and the temperature difference between the flue gases and the carbonising material is different at different periods of the treatment. The rate of heat flow towards the end of the treatment is therefore very small. · On the other hand, each part of a retort in a continuous process remains at the same temperature, and the retort may therefore be designed so that the exit temperature of the flue gases is continuously at a minimum. Incidentally the life of a retort working under steady conditions is likely to be longer than that of one working under intermittent conditions.

SIMPLICITY OF THE CONSTRUCTION AND OPERATION OF THE RETORT

To reduce liability to break down and the consequent maintenance costs, the machinery must be simple and as " fool-proof " as possible. In particular there should be a minimum of working parts, such as knuckle-joints, etc., exposed to high temperatures. Provision should be made for the expansion and contraction of the retort under varying temperatures. The plant should be capable of continuous operation for lengthy periods, and not require frequent attention for small repairs. It is desirable that control of temperature should be possible at various

parts of the retort, and local overheating of the material should be carefully guarded against. The retort should be air-tight as the presence of air not only destroys the coke but dilutes the gases and spoils the oil products. Leakage outwards increases the difficulty of controlling the temperature owing to the presence of an uncertain amount of combustible gases in the flues. The plant should require a minimum consumption of heat, power and water.

THE REDUCTION OF CAPITAL, MAINTENANCE AND LABOUR COSTS

In order to increase the financial rate of return on a plant of given size, it is obviously necessary that the capital expenditure and maintenance costs must be low, and that the throughput must be increased to a maximum.

Low capital and maintenance costs depend very largely upon the materials employed in construction and upon the general design of the retort. A plant which is bulky for the throughput attainable has less chance of commercial success than one of small size. Much of the by-product recovery plant is common to all processes and is now largely standardised, hence the costs of any proposed process may be appraised in terms of the costs of the retorting plant. Considerations affecting these will be discussed later.

The question of the best throughput for a given size of retort is one which is not easily decided. Experimental evidence is required to determine the best rate of rise of temperature of the carbonising mass. Speed is of decided importance in increasing the throughput, but throughput may be bought dearly if the material is only partially carbonised, while the effect of "stewing" at the maximum temperature on a commercial scale may have the result of decreasing the quality and quantity of the oil yield. The published data of gasworks experience, and the procedure in connexion with the shale oil industry of Scotland, are of great value, but this information must be modified to suit the different conditions of working for low temperature carbonisation, and further data from a commercial plant are required.

The size of a unit retort has also an important bearing upon the capital cost. The lower the throughput per unit retort, the larger must be the number of retorts for a given total daily throughput. There are certain advantages to be derived from having a large number of units, e.g. a stoppage in one retort for repairs or cleaning need not appreciably affect the working of the plant, etc., but there are also serious disadvantages such as the occupying of more ground space, more complications in charging and emptying, etc., all of which tend to increase the capital and maintenance costs. In developing a new process it is essential for the large-scale trial to be made on a unit plant of such a size as can be readily duplicated to form an economical battery should success be achieved. If, in order to reduce

the initial cost, work is done on a unit plant of appreciably less size than will be necessary in the full-scale plant, the results cannot be directly applied, owing to the differences introduced by the scale effect. It would appear that the throughput of a unit retort, built to try out a process, should bear such a relationship to the total throughput which is eventually to be carbonised that its multiplication into batteries would not render excessive the total number of retorts in any one bench.

The magnitude of the throughput of a plant has also an important bearing upon the maintenance costs. As the overhead charges remain nearly constant for all throughputs, it follows that the cost of retorting per ton is less for large throughputs than for small

Labour costs must also be low. The working operations should be simple, and the plant should require the minimum of attention after the final adjustments have been made. The process should preferably be continuous in action, so that labour-saving devices for charging and emptying the retorts can be easily employed. In installing labour-saving machinery, due regard must, of course, be paid to its capital and maintenance costs. A fuller discussion of the economics of the subject appears in the last chapter of this volume.

THE PRODUCTS OF LOW TEMPERATURE CARBONISATION

COKE

INTRODUCTORY

THE study of the behaviour of various types of coal carbonised at temperatures up to 600° C. or 700° C. has thrown much light on the influences which determine the physical structure and properties of the resulting coke. It has been shown that the sponge-like structure results from the simultaneous fusion and evolution of gas which occurs mainly between 350° C. and 700° C. It follows, therefore, that when coke is produced in ovens and retorts by the immediate application of external temperatures of 1200°–1400° C., the successive layers of the coal mass must necessarily pass through this stage of fusion and gas evolution. It may therefore be accepted that the ultimate structure of high temperature, as well as of low temperature coke, is mainly determined during this stage, and the study of what further occurs at higher temperatures, after the fusion stage is passed, can be more effectively pursued by direct application of these high temperatures to material which has been produced by the initial application of the temperature of 600° C.

A complete study of all varieties of coke is beyond the scope of the present volume. It will, however, be helpful to summarise at this stage some of the properties of cokes produced by high temperature processes, in order to see clearly the fundamental similarities and differences of high and low temperature products. The same course will be followed in the succeeding chapters dealing with the other products of low temperature carbonisation. ·

PRODUCTION OF HIGH TEMPERATURE COKE

It has already been stated that high temperature coke is produced in two existing industries, viz. (a) the gas industry, where the production of gas of suitable calorific value is regarded as the primary aim, but in which the solid residue usually represents the greater proportion of the potential energy of the coal carbonised, and (b) the coke oven industry, in which the chief object is to produce a dense, hard coke suitable for blast furnace or metallurgical uses, the tar and gas forming valuable by-products.

CHARACTERISTICS OF HIGH TEMPERATURE COKE

(a) GAS COKE

In the gasworks process carbonisation is carried out in externally heated horizontal or vertical retorts at temperatures ranging from 1000°–1300° C. The constitution, calorific value and other properties of the resulting cokes depend largely

upon the nature of the coals used in their production, since the whole of the ash remains in the coke, together with from 1 per cent to 6 per cent of volatile matter. The representative proximate analyses given in Table XXIV (p. 145) show that a gasworks coke has a composition of some 85 per cent "fixed carbon," 10 per cent to 13 per cent ash, 1 per cent to 3 per cent volatile matter, and about 1 per cent moisture. The coke for sale usually contains a higher percentage of water than this, and contents of from 5 per cent to 15 per cent, according to the conditions of quenching, storing, etc., are of frequent occurrence. Gas coke is a moderately hard, porous mass, with roughly rounded surfaces ; in colour it is greyish, with a somewhat metallic lustre, and frequently shows a yellowish tinge. It is softer than oven coke, and having been carbonised at lower temperatures, contains rather more volatile matter. The use of a strongly caking coal for gas making purposes in some parts of the country produces a coke which is sufficiently strong and hard to serve as a metallurgical coke, but as a general rule gas coke is unsuitable for this purpose. On account of the difficulty of ignition, and the unpleasant smell sometimes associated with its combustion, gas coke is not popular amongst domestic users. The difficulty of ignition is due partly to the low volatile content, partly to the high moisture content, and partly to inherent characteristics of structure of the coke.

CHARACTERISTICS OF HIGH TEMPERATURE COKE

(b) OVEN COKE

In the coke oven process the charges are firmly packed in recovery ovens and heated to temperatures of 1200° C. or 1300° C. The swelling nature of the coals used causes pressure to be exerted on the walls of the oven, and the final product is a very hard dense coke, which is capable of resisting heavy weights without crushing. It was clearly demonstrated by Dr. Lessing[1], in 1912, that the excessive frothing of certain coals could only occur when sufficient room was given for free expansion. If the coal is confined, the large bubbles, which would otherwise tend to form an excessively spongy and weak coke, are burst by contact with surrounding particles themselves confined by the walls of the retort. The effect is clearly shown in the laboratory apparatus devised by Lessing, in which ground coal is carbonised in a vertical silica tube heated externally. A loosely fitting plunger rests on the coal, preventing expansion, whilst at the same time allowing the gas to escape through the annular space between the plunger and the walls of the tube.

[1] Lessing, R. "A Laboratory Method for Comparing the Coking Properties of Coal." *Proc. Inst. Gas Engrs.*, 13th June, 1912. See also Fuel Research Board : Physical and Chemical Survey of the National Coal Resources, No. 2. Interim Report on Methods of Analysis of Coal. (H.M. Stationery Office.)

PRODUCTION OF LOW TEMPERATURE COKE

In the low temperature process, as its name indicates, the temperatures attained in the retorts, which are usually in the neighbourhood of 600° C., are much lower than those obtaining in either gas or coke works practice. In most of the present-day systems attention is directed chiefly towards making a coke with certain desirable characteristics not usually possessed by high temperature products. This can generally only be secured by leaving in the coke a greater proportion of volatile matter than would be left in high temperature coke, and the yield of gas is therefore low. It has already been explained that the fuel oil obtained is of great significance, and that the high calorific value of the gas makes up to some extent for its relatively small volume, especially in view of its value as an enriching agent for low grade gas.

There are essential differences in the various systems which have been adopted for low temperature carbonisation processes, but these fall approximately into three main classes, which are described in detail elsewhere ; in the first two, the retorts are externally heated, but in the third the crushed coal is exposed to the action of heat derived from producer gas, which is passed over and through the mass of coal. The nature of the cokes produced, both in regard to chemical consti-tution and physical characteristics, bears a close relation to the nature and mode of preparation of the parent coals, and to the particular method of carbonisation adopted, but in general low temperature cokes have a duller appearance than gas or oven coke, are softer and more friable, and are sometimes lacking in uniformity of texture. Their chief difference in chemical composition from gas or blast furnace coke lies in their relatively high proportion of volatile matter, which is usually between 7 per cent and 10 per cent. The temperatures employed in low temperature installations (up to say 650° C.) are so near the decomposition temperature of the coal itself, that the treatment which it receives inside the retort has a considerable influence upon the nature of the coke produced. As explained in Chapter X, non-caking bituminous coals give cokes which are soft and friable and quite unsuitable for transport, while the use of strongly caking coal renders difficult the satisfactory design of mechanical details of the retort. For satisfactory results, therefore, the coal used must either be sufficiently caking to yield a strong, coherent coke under the given working conditions, or must be mixed or blended with good caking coals in such proportions as to afford the necessary strength to the resulting coke. The results of recent researches have clearly indicated the important part which is played by the breaking down and blending of different types of coal in securing those qualities of homogeneity and physical texture which are so material to any form of smokeless solid fuel intended for domestic use. The effect, in addition, of briquetting fine coal by subjecting it to high pressures prior to

carbonisation is receiving much attention, and it is probable that this treatment will simplify and cheapen the subsequent operations of carbonisation. Experience has shown that the briquettes produced in this way from certain coal blends may lack robustness, and so tend to break down during the subsequent carbonisation ; but recent experimental work at H.M. Fuel Research Station has proved that by briquetting the same disintegrated and blended coals at temperatures high enough to cause incipient fusion of some of the constituents, a robust type of briquette may be obtained with a coarser material, and with a lower pressure than would otherwise be required.

VARIETIES OF LOW TEMPERATURE COKES OBTAINED FROM DIFFERENT PROCESSES

In Chapter XVII, details of the plant and methods employed in the various types of low temperature carbonisation processes will be given, but the compositions of certain varieties of low temperature cokes, which have been chosen as representing typical carbonising systems which differ from one another in some essential feature, are here given in Table XXIV, in conjunction with those of gas and oven cokes. Analyses Nos. 6 and 8, the Maclaurin and " Sensible Heat " (Nielsen) products respectively, represent cokes from internally heated retorts ; all the others are from externally heated retorts. The coke in Analysis No. 4 is that prepared at H.M. Fuel Research Station from a blend of caking and non-caking coals in such proportions that a slight shrinkage takes place on carbonisation, and no artificial pressure on the fused coal is necessary for the formation of hard coke. A coke of a similar character is that made by the Coalite process (Analysis No. 5), wherein coal blending is also employed, but where the fuel is in addition confined to some extent during carbonisation. In the " Pure Coal Briquette " process (Analysis No. 9) the coal briquettes are subjected to varying stages of heating as they pass down the retorts, and the method also differs from others in that the coke is finally heated at the bottom of the retort to temperatures which are high enough to reduce the quantity of volatiles to about 2 per cent or 3 per cent. As has already been stated, the authors of this process consider that the fine cell structure of the emergent coke, which will be discussed in more detail later, compensates for the lack of volatile matter, so that the free burning qualities of the cokes are not forfeited.

THE CHARACTERISTICS OF LOW TEMPERATURE COKES

(c) COHERENCY

The coherency of a coke has already been shown to depend upon the presence in the coal in sufficient quantity of substances which upon carbonisation act as

binding materials to other constituents. If this binding material is in excess, steps must be taken to prevent excessive foaming. This may be effected in various ways, such as by confining the coal, as discussed above, or by subjecting the material to a pre-heat treatment or to oxidation, which will destroy some of the binding material. The most promising method, however, is to provide, by blending with a non-caking coal, a sufficiency of more inert material to absorb and so utilise the excess binder in the swelling coal.

Figs. 15 and 16, reproduced by permission of Sir George Beilby, illustrate cokes produced from different coals in the laboratory assay apparatus. In Fig. 15, at A, is shown the result of carbonising a type of coal (Ellistown Main Breeze) deficient in caking power, the residue taking the form of a non-coherent powder only suitable for briquetting or burning in the pulverised form. On the other hand, at C, the coke from Mitchell Main coal has foamed up during carbonisation and become stereotyped in that form. It is friable and quite unable to stand transport. At B the result is shown of intimately mixing the above two coals before carbonisation. The coke, which has shrunk during the process, is strong and fine grained and eminently suitable for use as a smokeless fuel.

It is well known that most coal seams present a succession of bands of variable fusibility, each of which if carbonised alone would produce a coke of different structure. It will be seen therefore that such seams, when carbonised as a whole, will produce cokes of varying characteristics according to the proportions of the caking or non-caking bands present. It sometimes happens that the two types are present in such proportions that a good coke is produced without additional blending or external pressure. An example of such a coke from Dalton Main coal is shown in Fig. 16, A being a side view and cross-section, while B is a plan view of the same specimen.

Messrs. Sutcliffe and Evans[1] claim that most coking coals briquetted cold with 25 per cent of breeze without a binder and subsequently carbonised will give an excellent smokeless fuel, but it must be borne in mind that the breeze in re-carbonising takes up space in the retorts, and yields no gas. In the carbocoal process, low temperature coke is ground, blended with pitch and briquetted, and the briquettes carbonised until a low volatile product is obtained. The carbocoal briquettes are dense and strong and form an efficient substitute for anthracite.

(b) VOLATILE CONTENT

Table XXIV shows that the possession of 7 per cent to 10 per cent of volatile matter is a common property of low temperature cokes other than the exceptions referred to above. This high volatile content is to some extent responsible for the

[1] Sutcliffe and Evans. "Low v. High Temperature Carbonisation." *Proc. S. Wales Inst. Engrs.*, 20th April, 1922.
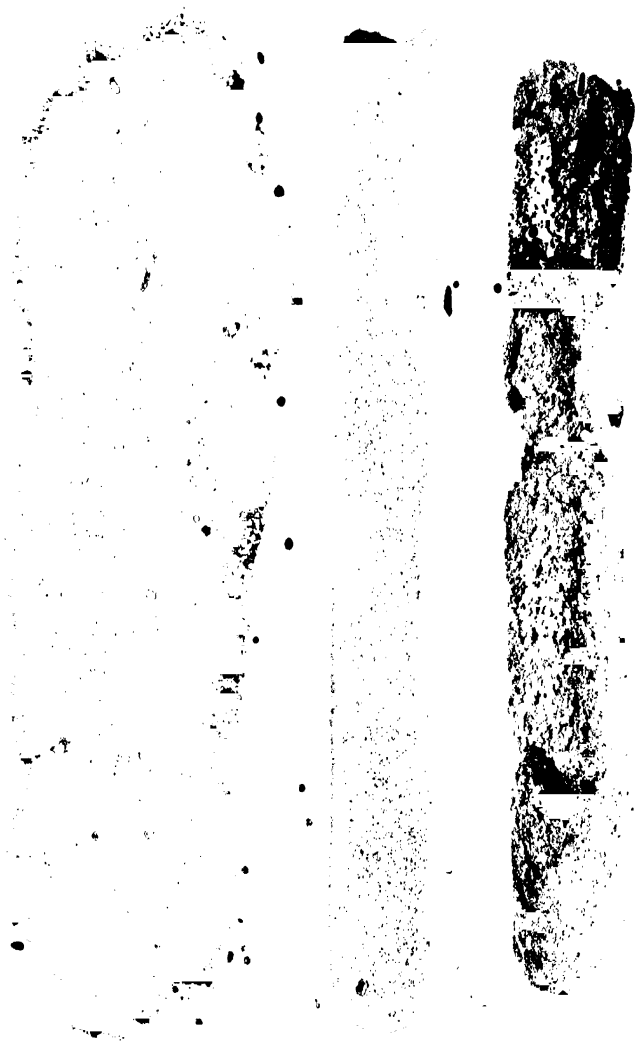
FIG. 15.—LABORATORY ASSAY COKE, SHOWING THE RESULTS OF BLENDING COALS OF WIDELY DIFFERING FUSIBILITY.

a.—The anthraceous *a*-side from Ellistown Main breeze is practically a non-coherent powder.
b.—The coke from a blend of 40 per cent. of Ellistown Main with 60 per cent. of Mitchell Main is relatively hard and dense and occupies a much smaller volume than the original coal.
c.—The coke from Mitchell Main is an open sponge.

TABLE XXIV.

ANALYSES AND CALORIFIC VALUES OF DIFFERENT TYPES OF COKE.

TYPE OF COKE.	GAS COKE.			LOW TEMPERATURE COKES.						
	1	2	3	4	5	6	7	8	9	10
	Coke Ovens.	Horizontal.	Vertical.	H.M. Fuel Research Station.	Coalite (Barnsley).	Maclaurin Process.	Carbo-coal.	"Sensible Heat" (Nielsen) Process.	"Pure Coal Briquette" Process.	Tozer.
Proximate Analysis:—										
Moisture	3·0	1·53	0·62	0·48			1·84	1·90	4·53	
Volatile Matter (less moisture)	0·5	2·82	0·88	8·35	10·0	4·77	2·75	11·50	3·66	12·27
"Fixed Carbon"	86·5	85·89	85·10	79·30	83·6	86·71	85·64	70·70	82·48	75·45
Ash	10·0	9·71	13·40	11·87	6·4	8·52	9·77	15·90	9·33	12·26
	100·0	100·00	100·00	100·00	100·0	100·00	100·00	100·00	100·00	100·00
Ultimate Analysis (dry):—										
Ash	10·3	9·86	13·48	11·93				16·20	9·78	
Carbon		87·55	83·57	77·87				72·50	83·27	
Hydrogen		0·38	0·48	2·57				3·00	1·05	
Sulphur (comb.)	1·0	0·66	0·99	1·49				1·80		
Nitrogen		0·99	1·34	1·43				1·50		
Oxygen (diff.)		0·56	0·14	4·71				5·00		
		100·00	100·00	100·00				100·00		
Calorific Value, B.Th.U. per lb.:—										
Corresponding to Prox. Anal.		12,590	12,160	12,960		12,660	13,080	11,500	12,285	
Ash-free Dry		14,200	14,130	14,790		13,780	14,780	14,000	14,270	

(1) C. A. Meissner. Iron and Steel Inst., U.S.A.
(2) Horizontal Retort. South Met. Gas Co. Analysis at H.M. Fuel Research Station.
(3) Vertical Retorts. H.M. Fuel Research Station.
(4) Horizontal Low Temperature Retorts. H.M. Fuel Research Station. Consett Coal, 20 per cent steam. F.R.B. Report, 1920-21. 2nd section.
(5) Engineering, October, 1921, 112, 598.
(6) McLaurin, Journ. Soc. Chem. Ind., 1917, 36, 620.
(7) Brownlie, Industrial India, II, No. 9, p. 479.
(8) Engineering, 1922, 113, 347.
(9) Analysis at H.M. Fuel Research Station.
(10) F. M. Perkin, Journ. Soc. Chem. Ind., 31.7.1918.

easy ignition and free burning properties of such cokes, and it has been shown at H.M. Fuel Research Station that the same process, in conjunction with different times of carbonisation, produces cokes of different combustibility, the easy combustibility of cokes containing volatiles of the above order being to some extent lost if the time of carbonisation is so prolonged as to reduce the volatiles to less than 4 per cent. Bünte and Kolmel,[1] however, claim that a low temperature coke once produced can be heated to increased temperatures out of contact with air, until all the volatile matter is removed, without interfering with its property of ready ignition.

<div align="center">(c) STRUCTURE</div>

It would appear, therefore, that properties other than chemical have a considerable bearing upon the combustibility of a coke. Recent experimental work has indeed amply demonstrated the important consequences of structure, and has shown that one of the main factors involved is the area of the cell surfaces upon which gases can act, and which it is desirable should be as large as possible. The structure of a coke is determined to some extent by the nature of the carbonising plant used, but is chiefly influenced by the action of the caking constituents of the parent coal. The results of the ordinary methods of coal analysis may serve for a rough classification of coals into types, but very little information is actually gained thereby as to the real nature of the coal, or its probable behaviour on carbonisation. Further, most coal seams are built up of bands of coal which individually may differ considerably from the seam average, and which, therefore, if carbonised alone, would give correspondingly different results. The temperature of carbonisation and the conditions under which it is carried out also affect the physical properties of the coke product.

The question of the structure of coke was reviewed by Sir George Beilby in 1922,[2] when he described certain variations in structure of varieties from wood charcoal to metallurgical coke. In wood charcoal the original cell structure of the wood is preserved, but observation showed that in coke produced from coal the final cell structure is that of bubbles formed from fused or partially fused coal substance. The first stage will be the formation of a foam in which each bubble is a self-contained cell, but mutual perforation of points of contact produces a spongy mass through which the gases ultimately escape. As the temperature of the coal mass is raised, gas constantly forms; the development, therefore, is continuous. Bubbles are formed so long as the mass remains sufficiently plastic; but when this stage is passed no further development can take place, although gas may still be evolved without further modification of structure. The temperature at which carbonisation

---

[1] Bünte and Kolmel, "Coke Ignition Temperatures." *Gas u. Wasserfach*, October, 1922.

[2] Beilby. "The Structure of Coke. its Origin and Development." *Fuel in Science and Practice*, December, 1922.
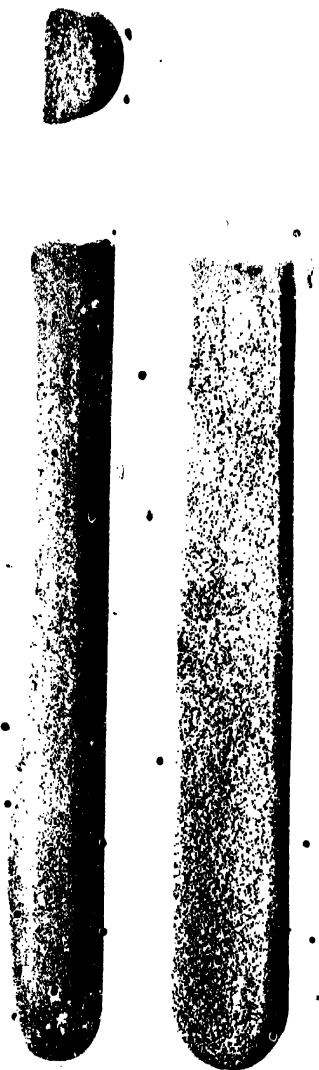
FIG. 16.—COKE PRODUCED IN THE LABORATORY ASSAY APPARATUS (NATURAL SIZE).

A and B—Dalton Main, a typical coal which produces a satisfactory coke for use as smokeless fuel for domestic purposes. Compare with Fig. 23.

takes place, or, rather, the rate at which the temperature of the coal is raised, has a considerable bearing upon the final structure of the coke which has been attained when rigidity sets in. Unless confined in some way, such as, for example, by solid boundary surfaces, the large bubbles which will be blown in a swelling coal will result in the production of a light, porous mass. If the coal, or coal blend, contains the correct proportion of fusible constituent, the final bubble structure will be governed by the initial state of division of the particles. With finely divided coal, for example, it will consist of minute cells of uniform structure. A dense fuel, with a large number of such small cells per unit of the vitreous carbon mass, is the ideal generally aimed at. It has been successfully attained, as already mentioned, by the blending of fusible and non-caking coals, which is fully described in the Report of the Fuel Research Board.[1] In this method, mixtures can be obtained which will pass through the fusion and bubble-forming stages without external swelling, and which will yield a fairly hard coke of slightly less volume than the original coal.

Several other investigators have shown that a similar effect can be produced by using coke breeze instead of the non-caking coal, and in many other low temperature carbonisation processes it is now recognised that coal blending is a very satisfactory method of giving a suitable fuel. In the " Pure Coal Briquette " process already referred to, the coal is very finely ground after blending, in order that the ultimate structure of the coke may consist of very minute, but interconnected cells. In this connexion it is of interest to note the high apparent density of this type of coke, which has been given as 1·15 in comparison with the more usual values of 0·7 to 0·9 for other varieties of low temperature coke.

(d) DENSITY, POROSITY AND COMBUSTIBILITY

Sir George Beilby, from his studies of the formation of carbon by the carbonisation of sugar, and of the microstructure and properties of coke, has arrived at the conclusion that most carbonaceous residues, from wood charcoal to the hardest coke, consist of carbon in the vitreous state, i.e. it is a true glass. The density of this material is in the neighbourhood of 1·85 to 1·9 as compared with 3·55 and 2·55 for diamond and graphite respectively. Until recently, metallurgists have considered density and porosity to be reciprocal the one to the other, that is to say, the usual method of calculating porosity has been by means of the ratio of the relative volumes of actual coke substance and interspaces. It is obvious that a value for porosity calculated in such a manner can give no indication of the actual area of internal surfaces developed ; neither can any opinion be formed as to the accessibility of such internal surfaces to an oxidising or reducing agent. Combus-

[1] Fuel Research Board. Report for Years 1920, 1921 ; Second Section : "Low Temperature Carbonisation." (H.M. Stationery Office.)

tibility has been generally regarded as depending on chemical rather than structural features in the coke; but if the actual material of cokes possessing widely differing degrees of reactivity is such a definite substance as vitreous carbon, we must look elsewhere for an explanation of the differing properties. The most likely direction is obviously in the amount of surface exposed to chemical action. A similar mass of carbon can obviously differ very widely in this respect. For example, we may have a solid mass of vitreous carbon concentrated in a cube of say one inch side, so that the area exposed to oxidation is six square inches, presuming a similar degree of exposure in each of the six surfaces of the cube. Again, the same weight of material might be arranged in the form of cubes of $\frac{1}{10}$-inch side, with spaces of say $\frac{1}{10}$-inch between the faces of the cubes. The area of each face of each cube would now be $\frac{1}{100}$ of a square inch, or a total of $\frac{6}{100}$ square inches to each cube. But, since we now have 1000 cubes, the total surface would be $\frac{6}{100} \times 1000 = 60$ sq. inches, or ten times as much surface as was possessed by the original 1-inch cube. Further subdivision would result in increasing the surface area to any extent we please.

In the original cube, reaction can only take place over the external surface; but in the modified systems reaction can proceed on the surfaces in the interior, provided always that a circulation can be set up which shall furnish a sufficient supply of oxygen for combustion, and at the same time remove efficiently the products of combustion. However, if we proceed as above to increase the surfaces, we are at the same time rendering them more inaccessible to the action of the other element necessary to the reaction. It would therefore appear that the increase of surface provided by minute subdivision will be eventually neutralised by difficulty of access of air to the interior. It is obvious, therefore, that the usual method of estimating porosity can give no indication of the relative area of the internal surfaces, neither can it give any idea of the accessibility of such internal surfaces. In this connexion Sir George Beilby has determined the sizes of the larger bubbles in a typical metallurgical coke, since these are likely to be the most accessible. Such bubbles range from 3 or 4 mm. down to 0·5 mm. in diameter, and he considers that the "free circulation of gases will mainly occur through the larger of these, as they have probably been opened up and kept open by the rapid discharge through them of the carbonisation gases."

For dense, pre-briquetted coke obtained by fine grinding, he has estimated that the larger pores range from 0·5 mm. down to 0·1 mm., but points out that "the fact that the combustion of these briquettes proceeds definitely from the outer surface inwards shows that the internal circulation of the oxidising gases is much more restricted than in the case of metallurgical coke. Sutcliffe and Evans believe that the reactive surface of the coke briquette is so immensely greater than that of metallurgical coke that the increased rate of combustion more than compensates for the less free circulation through the mass."

FIG. 17.—GAS RETORT COKE.   FIG. 18.—METALLURGICAL COKE.

FIGS. 17 AND 18.—COKE STRUCTURE. REPRODUCTION OF MICROGRAPHS AT A MAGNIFICATION OF 50, WHICH WERE PUBLISHED BY DR. THÖRNER IN "STAHL UND EISEN" IN 1886.

Sections of coke were prepared by grinding and were photographed by transmitted light. As the thickness of these sections, unlike those which can be prepared from coal, cannot be so far reduced that they become transparent, the resulting pictures are simply silhouettes produced by the passage of light through holes in an opaque screen. There is not even any certainty that the holes really represent the size or shape of the pores or elements of structure, for in so fragile a material as vitreous carbon in the form of thin films the grinding of the section might easily lead to the destruction of the more minute elements.

From the above it is obvious that a coke may burn either entirely at the external surface or partially externally and internally at the same time, according to its structure. Cokes of both types have been produced under low temperature conditions at H.M. Fuel Research Station, and both present attractive features. In the coke cakes, made in the horizontal retorts at temperatures of 600° C., the burning appears to take place at the surface and for some distance into the interior, and this certainly gives the most attractive fire.

Low temperature coke, formed by preheating disintegrated coal to a temperature at which incipient fusion takes place, and briquetting at this temperature, followed by carbonising at 600° C., appeared to burn mainly at the surface. The carbonised fuel is very dense, and as strong as a good metallurgical coke, but on burning deposits a fine ash on its surface, which, if not brushed away, produces after a time a rather dead and unpleasant-looking fire. In the authors' opinion the best result will probably be obtained by using coarser particles for briquetting, so that if a sufficiently robust briquette can be obtained to stand the subsequent carbonising treatment, a more open structure will be evolved, which will at the same time give a fuel denser than the horizontal retort coke cakes. This, however, remains to be proved, and the relation of "accessibility" of surfaces to rate of combustion is now being investigated both at H.M. Fuel Research Station and elsewhere.

The microphotographs, reproduced by permission of Sir George Beilby in Figs. 17 to 30, illustrate the appearance at different (low) magnifications of cokes ranging from wood charcoal to blast furnace coke. Beautiful as these photographs are, Beilby has pointed out that they cannot take the place of patient microscopic study of the actual specimens, using lenses of gradually increasing resolving power. The necessity for such study to the proper appreciation of the arguments in Beilby's paper strongly impressed itself upon one of the authors, who had the privilege of following Beilby's work in this connexion, stage by stage as it was developed.

FORMATION OF COKES UNDER LABORATORY CONDITIONS

The laboratory assay method, which has been elaborated at H.M. Fuel Research Station, has proved particularly useful for the study of different types of coke produced by carbonisation of coal at low temperatures, and by its means the influence of the fusibility of the coal upon the ultimate structure of the coke can, to some extent, be followed. It was possible at the Research Station to correlate these results with those obtained on a larger scale in a setting of horizontal retorts which had been specially built for low temperature carbonisation experiments. In the preliminary work which led to the perfected methods of test, it was observed that the time of heating exerted a marked effect upon the final structure of the coke. On the small scale of the assay apparatus (20 grammes) immediate sub-

jection to the full temperature of 600° C. resulted in a very much more swollen coke than if the mass was heated more gradually. ' Heating of the laboratory assay tube by stages was soon discarded, as it led to condensation of the tar vapour along the charge and its consequent secondary decomposition. Attention was therefore directed towards finding a set of conditions which would obviate so far as possible decomposition of the primary gas and tar, and yet produce a coke which was similar to that formed from the same coal in large-scale practice. The most important factor was obviously the rate of heating, and since on a large scale the rate of heating is much slower than in any laboratory apparatus, it was decided to slow down the rate of heating in the assay method by starting at a low temperature and working up to the final. In an intermittent retort the outer layer is heated rapidly to nearly the full temperature, while the centre of the charge is probably not heated through for several hours. In a vertical continuous retort the rate of heating would be more uniform, and the coal may be regarded approximately as passing through successively increasing zones of temperature to the final carbonising zone. The condition finally chosen for the assay was that the retort containing the dry coal should be introduced into a chamber at '300° C., or well below the point of visible decomposition, and the temperature raised during one hour to the full value desired. A further hour at this temperature completed the carbonisation.

The cokes obtained in this way were very similar in appearance and yield to the horizontal retort cokes, and when at a later stage a coke which would bind well without swelling was sought for, the apparatus became extremely useful for predicting the necessary proportions of caking and non-caking coals.

LOW TEMPERATURE COKE AS A HOUSEHOLD FUEL

The most important potential use for low temperature carbonisation coke is undoubtedly as a domestic fuel, though tests have shown it also to be well adapted for industrial purposes, e.g. a gas producer fuel. The open domestic fire, since it attains a relatively high temperature, emits the greater part of its energy as radiation. Determinations of the heat radiated therefore form a reasonable basis of comparison of the behaviour of different fuels for domestic purposes. The work of Dr. Margaret Fishenden[1] has shown conclusively that the radiating power of coke, especially that produced from low temperature processes, is greatly superior to that of raw coal; for instance the radiation efficiency, or the proportion of the theoretical energy of combustion of the fuel burned, delivered as radiation in three different types of sitting-room grates, was found by Dr. Fishenden to be 24·2, 19·5, 17·3 respectively for bituminous coal, and 30·8, 24·0, 19·9 for a low temperature

[1] Fuel Research Board. Tecn. Pap. No. 3. "The Efficiency of Low Temperature Coke in Domestic Appliances." Margaret White Fishenden, D.Sc. (H.M. Stationery Office.)
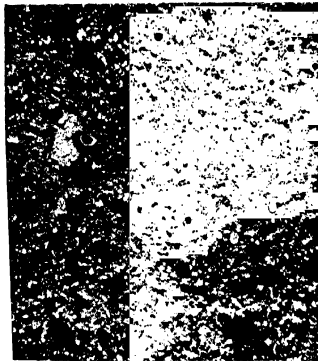
Fig. 19 is gas coke from horizontal retort using Durham coal.

Fig. 20 is blast furnace coke from recovery ovens. Compare with that shown in Figs. 17 and 18.

Fig. 21 is a cross-section of laboratory assay coke from Mitchell Main, a fairly fusible coal. Considerable frothing has occurred during carbonisation. Compare with Fig. 15.

Fig. 22 is gas coke from vertical retorts using Arley, a fairly fusible coal. Compare with the enlargement of this, Fig. 25.
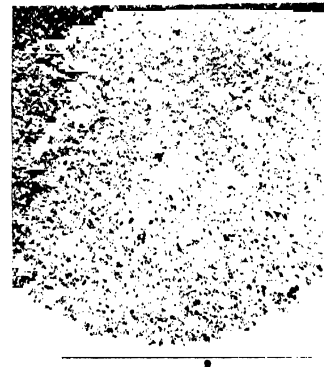
Fig. 23 is a cross-section of laboratory assay coke from Dalton Main. Compare with Figs. 16a and b.

Fig. 24 is the coke resulting from the carbonisation of a briquette made by the compression of pulverised coal. The structure is one, imperfectly resolved by the planar lens. Compare Fig. 26.

FIGS. 19–24. COKE STRUCTURE.

Photomicrographs by Sir G. Bailey in 1922. These were obtained by the use of a planar lens of 3-inch focus stopped down to give depth of focus. The illumination was by reflected light at an angle of about 45°. The magnification was from 5 to 10 diameter.

coke. The comparative performance of the two fuels, weight for weight, will obviously be dependent upon the relative calorific values, but it is apparent from the figures quoted that a low temperature coke of calorific value, say, 12,500 B.Th.U. per pound, will in general give considerably more radiation than a coal fire burning at the same rate. It will be noticed also in the figures quoted above that the higher the efficiency of the grate, the more pronounced was the advantage of the carbonised fuel over raw coal.

In addition, the easy ignition and free burning properties of low temperature cokes, the bright, hot, glowing and extremely attractive fires which they produce, and the entire immunity of these fires from black smoke emission, render low temperature cokes a clean and convenient household fuel ; and if such cokes became available in large residential centres the problem of increasing the efficiency of the open fire would be greatly simplified.

When raw coal is burned in the open fire, its distillation *in situ* is the cause of the smoke trouble. The smokeless combustion of the products of this distillation—gases and tar—can only be obtained by continual care and attention that cannot be given in practice. Hence the chimney of an open fire which is continuously used must be swept several times yearly, while the unpleasant task of cleaning dampers and flues must be undertaken every few days. With smokeless solid fuel, on the other hand, the operation in the fire is simply one of combustion, and no regulation is required other than that of the air supply, which controls the rate at which the fuel burns. On account of the absence of smoke production, chimneys of reduced size are adequate, and corresponding economy is gained in building and maintenance costs. The draught regulation itself is also simplified by the more uniform action of flues and dampers, which remain clean and unblocked by soot or tar.

For water heating, especially in independent boilers, low. temperature cokes are again greatly superior to coal in actual efficiency, values of 20·7 per cent and 17·3 per cent having been obtained by Dr. Fishenden from the back boilers of ordinary kitcheners as compared with corresponding figures of 14·5 per cent and 13·1 per cent for coal ; and in an independent boiler an efficiency of 41 per cent for low temperature coke contrasted well with 31 per cent for coal. Their easy ignition, freedom from smell, and certainty and simplicity of regulation differentiate them favourably from gas coke.

For oven heating in ordinary kitcheners, low temperature coke appears to be somewhat less advantageous than raw coal. In most of the tests about equal numbers of heat units were required roughly to give similar results in oven heating or to cook similar menus. In certain special ranges based upon independent boiler design, however, low temperature cokes appear to be better than coal.

The probable behaviour of these brightly glowing fuels, with low volatile

contents, and consequently little flame, may frequently be anticipated from the design of the appliance in which it is proposed to burn them. Their advantage, in fact, is largely due to enhanced radiation—though the non-luminous flame produced from the remaining volatiles is not without its effect—and is most marked when the distance between the fuel bed and the oven or boiler surfaces is low, or the fuel bed enclosed. .Where the oven or boiler surfaces are far removed from the fuel bed, the superiority of coke is diminished, or even in certain cases may disappear. In these cases the direct contact of the long and far-reaching flames of bituminous coals apparently more than counterbalances the higher radiation from the coke. When it is considered that the practical comparisons given above refer generally to appliances planned to burn coal, the favourable results obtained with the new fuels may be considered highly satisfactory, for there is little doubt that in appliances specially designed for the latter, their superiority might be considerably magnified.

For household purposes, it is essential that cokes should be produced in such a form as to be easily handled. Moreover, the individual pieces should be roughly uniform both in size and structure, so as to be suitable for charging on any ordinary domestic grate. They should also be strong enough to resist severe handling without excessive breaking, and be reasonably clean to touch. In the early days of low temperature carbonisation, the resulting cokes from the non-caking coals used were friable and uneven in structure ; but the material produced more recently, if not entirely free from these faults, proves, both in size and shape, a highly convenient fuel. No one who has had experience of the bright, hot, uniform and extremely pleasant fires which good low temperature cokes produce would wish to exchange them for the old, smoky coal fire, and the economic production of a satisfactory low temperature coke on a large scale should prove an immediate amelioration of the smoke nuisance.

LOW TEMPERATURE COKE AS A PRODUCER FUEL

Low temperature cokes are eminently suitable for gas producers, as the gas formed is higher in calorific value 'than that from gas coke, and is free from the tarry deposits characteristic of richer fuels. The practical application of low temperature coke to gas producers offers a means of making a very ready comparison of its increased combustibility over that of ordinary gas coke. Table XXV gives the results of experiments carried out at H.M. Fuel Research Station on two samples of coke crushed to the same size, and burned in a small gas producer under identical conditions. It will be seen that the rate of combustion of carbon was 23 per cent higher for the low temperature coke than for the gas coke. The greater speed of reaction is also indicated by the increased temperature (200° C.) of the hot zone of the producer in the case of the low temperature coke.

FIG. 25. ENLARGEMENT OF FIG. 22. COKE FROM ASTLEY COAL
CARBONISED IN VERTICAL RETORTS WITH STEAMING

The continuity of the bubble sponge structure can be followed on this photograph from cells of 1 mm. down to 0·05 mm. diameter. The more minute cells in the walls of the larger bubbles, which appear only as small diffraction discs, when visually resolved by the 16 mm. apochromat show the continuity of this structure down to a minuteness of 0·001 mm.

A somewhat similar method of comparing the combustibility of cokes has been described by G. Tammann.[1] In this method the cokes are consumed in small cylinders, open at the top, into which equal quantities of air are blown. The relative combustibility is then measured by comparing the heights of the cones of burning carbon monoxide.

TABLE XXV

COMPARISON OF LOW TEMPERATURE COKE AND GAS COKE USED IN PRODUCERS

	H.M. Fuel Research Station : Low Temperature Coke.	Vertical Retort Gas Coke.
Rate of consumption of coke : lb. per hour	8·63	6·12
Rate of consumption of carbon : lb. per hour	6·56	5·33
Temperature of hot zone	1000° C.	800° C.
Composition of gas made :—		
CO_2	10·1	14·3 -
O_2	0·4	0·9
CO	17·2	8·6
H_2	——	——
CH_4	—	—
N_2	72·4	76·2

LOW TEMPERATURE COKE AS AN INDUSTRIAL FUEL

Considerable attention has been devoted during recent years to the application of gas coke as a steam-raising fuel partly on the grounds of smoke abatement, and partly with a view to absorbing the continually increasing coke yields of our gas-works. For these reasons many large concerns have adopted coke as a boiler fuel. The Paris Gas Company in 1920 adopted gas coke for boilers, after carrying out for one year tests in which gas coke and coal were found to be about equivalent, weight for weight, the higher efficiency given by the coke, and the reduced loss in small particles of unburned fuel, making up for the lower calorific value. The value of the pioneering work of Mr. E. W. L. Nicol[2] and the London Coke Committee, both during and since the war, in encouraging the industrial use of gas coke, is of great importance, and is discussed at length in another volume of the present series.

Although there are few published data regarding the efficiency of low temperature coke as a fuel for steam raising, such experience as is available shows that it shares the advantages of coke firing without some of the disadvantages associated

[1] G. Tammann. *Stahl und Eisen*, 1922, 42, No. 15, pp. 577–8.
[2] Nicol, E. W. L. *Coke and Its Uses.* (Benn Bros.)

with the high temperature products. Low temperature coke as a boiler fuel behaves in a similar manner to a smokeless Welsh steam coal. It is easily kindled and burns freely with a good radiation efficiency. The total absence of smoke-producing volatile matter renders it easy to maintain the chimney absolutely smokeless. Its only disadvantage is that associated in some cases with its bulk, so that weight for weight with hand stoking, the firing has to be more frequent than in the case of coal, in order to maintain a uniform fire and so obtain the maximum economy. The pre-briquetted form of low temperature coke being much denser would of course not share this disadvantage.

PULVERISED LOW TEMPERATURE COKE

The possibility of using low temperature coke in the powdered form has been pointed out by S. W. Parr and C. K. Francis.[1] The burning of fuel in the finely divided form has lately been making great strides in the United States of America and on the Continent. In this country, however, with the exception of the cement industry, where powdered coal is used exclusively in the rotary kilns, progress in the utilisation of pulverised fuel has been disappointing. Signs are not wanting, however, that our industrialists are beginning to realise that there are certain manifest advantages to be gained by the utilisation of fuel in this form. Progress is likely to be hastened by the most recent development which has taken place, viz. the pulverisation of the fuel in small self-contained pulverisers, situated practically at the furnace mouth. This method completely obviates the necessity for the cumbersome and costly plant associated with some of the earlier installations.

The advantages of pulverised fuel are summarised in the *General Electric Review* for May, 1918, as follows :—

(1) Flexibility of control of fuel and air, and ability to extinguish the fire instantly.

(2) Complete combustion even at high rate of burning, and elimination of smoke, always assuming that the installation is properly made and operated.

(3) Burning fuel in suspension eliminates the usual troubles which result from the formation of clinkers in the fire bed when the coal is burned in grates.

(4) Low grade fuels may be burned efficiently regardless of the proportion of ash, sulphur or other impurities. Where low grade fuels are burned in grates, the capacity of the furnace is reduced in proportion to the percentage of incombustible content. This limitation does not hold when burning pul-

---

[1] Parr, S. W., and Francis, C. K. "The Modification of Illinois Coal by Low Temperature Distillation." *Bulletin*, 24, Univ. of Illinois.

Fig. 27 is vitreous carbon prepared from sugar. Though large bubbles, both burst and unburst, are seen, the more minute structure seen in coke is absent.

Fig. 28 is the broken surface of a briquette made from pulverised coal (about 60 mesh) heated to 380 C. and compressed at 2 tons per sq. in.

Fig. 26 is coke resulting from the carbonisation of the briquetted blend of Mitchell Main and Ellistown Main at a higher temperature than that employed in Fig. 24.

Figs. 29 and 30 show oak charcoal across and along the grain of the wood. The structure is practically a reproduction of the natural cell-structure of the wood. There is little or no evidence of fusion and bubbling.

FIGS. 26–30.—COKE STRUCTURE.

Photomicrographs by Sir G. Beilby in 1922, prepared under the conditions described for Figs. 19–24. Magnification 5–10 diameters.

verised fuel in suspension, as the amount of ash in suspension in the flame at any one time is inconsiderable.

(5) Very little excess air is required. This reduces the stack loss as well as the power required for the draught blowers. Less area is also required in flues and stacks.

(6) Maximum fuel economy is possible in many applications.

(7) The expense of supplying coal to scattered industrial furnaces is thereby reduced to a minimum. Pulverised fuel has semi-fluid properties ; it flows easily and can be transferred through pipes,

(a) by screw conveyors ;
(b) in a mass by means of compressed air ;
(c) in suspension in a current of air.

For satisfactory and easy burning in the pulverised form it is desirable that a fuel should contain about 10 per cent of volatile matter. Since this is the amount usually left in a low temperature coke, pulverisation is a very attractive method for utilisation of the fines and breeze which are associated with some of the systems. Indeed, if there were a free market for the fines such as would present itself if there is in the near future a great development of powdered fuel installations, the range of coals available for low temperature carbonisation would be widened, and certain early types of retort might become profitable in working.

Another advantage of low temperature coke as a raw material for pulverisation is its comparative friability. One of the chief difficulties encountered in the pulverisation of some other fuels is the excessive wear and tear of the grinding apparatus. It is unlikely that it would be economically sound, unless in exceptional cases, to run a carbonising plant for the purpose of disposing of the whole of its coke as a material for pulverisation ; but, if it were possible to do this, the carbonising plant and the coal carbonised could be arranged to deliver a coke in which the property of easy pulverisation could be developed to the utmost extent.
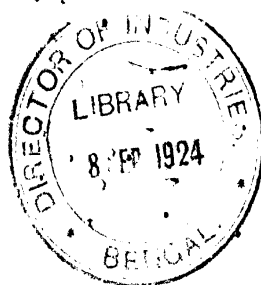
THE PRODUCTS OF LOW TEMPERATURE CARBONISATION

GAS

COMPOSITION OF GAS OBTAINED FROM LARGE-SCALE PLANTS

It has already been explained that the composition of the gases which are evolved during the thermal decomposition of bituminous coal varies not only with the temperatures employed, but also with the time over which the operation extends, the rapidity with which the volatile products emitted are removed from the retorts, and other practical conditions.

In any large-scale practice, the primary products of distillation are subjected for some time to the heat of the retort. Owing to the secondary decomposition which is thereby caused, neither the gaseous nor liquid products of carbonisation actually collected represent the true products initially liberated from the heated coal. Moreover, as coal is an indifferent conductor of heat, the temperature of the charge in the retort is far from uniform, for while certain parts may become heated rapidly, others pass through a more gradual change.

It is not therefore possible to give specific analyses of the gaseous products associated with different coals and different temperatures of carbonisation which will apply precisely in all cases. The published data of large-scale gas compositions are few, but Table XXVI shows analyses of the actual gases obtained in two large-scale plants which are typical of external and internal methods of heating. These are : Fuel Research Board horizontal retorts and the Maclaurin retort. The former may be compared with the composition of the gas produced on the laboratory scale by Wheeler, using Altofts Silkstone coal, carbonised at temperatures of 450° C. and 600° C. respectively, and that obtained from H.M. Fuel Research Station Assay method already described. (Page 60.) The gas obtained with the internal system of heating is not comparable with these laboratory results since it contains combustion and producer gases inherent to its particular system of heating.

COMPOSITION OF GAS OBTAINED IN LABORATORY EXPERIMENTS

In Tables XXVII and XXVIII detailed figures of Burgess and Wheeler's[1] small-scale experiments in vacuo at temperatures ranging from 100–800° C. are shown, and bring out clearly the critical period in the decomposition of the coal between 700° C. and 800° C., which has already been discussed in Chapter VI.

In general the composition of the gases obtained from full-scale installations differs considerably from that yielded in laboratory experiments, though that produced in the large-scale experimental plant at H.M. Fuel Research Station is closer than many others. The causes of the divergence may be classed broadly as (1) the dilution

[1] Burgess and Wheeler. "The Distillation of Coal in a Vacuum." *Trans. Chem. Soc.*, 1914, **105**, 131–40.

of the rich gas made with a heating gas or a gas made at a different temperature, and (2) the "cracking" of the rich gas and primary tar to a greater or less degree. The Maclaurin retort, in which the heating is produced by the internal combustion

TABLE XXVI

COMPOSITION OF GAS MADE IN PROCESSES FOR THE CARBONISATION OF COAL AT LOW TEMPERATURES

Process.	Burgess and Wheeler[1] (Lab. scale).		H.M. Fuel Research Stn.		Maclaurin Retort.[2]
			"Assay"	Horizontal Retorts.	
Retort temp. ° C.	450	600	600	600	700
Coal used	Altofts Silkstone		Dalton Main		
Gas made c. ft. per ton of dry ash-free coal	431	3550	3680	3440	27,730
Analysis :—					
CO_2, H_2S, etc.	15·65	4·90	8·8	7·6	6·2
C_nH_m	9·45	7·00	3·6	9·1	Nil
O_2	—	—	—	—	0·6
CO	8·75	7·10	5·8	5·8	16·0
H_2	7·00	26·60	26·3	17·1	16·1
C_nH_{2n+2}	59·10	54·40	55·4	56·0	13·0
N_2	—	—	—	4·4	48·1
Value of " n " in paraffins	1·57	1·35	1·27	1·24	—
Calorific value. B.Th.U.'s per cu. ft. @ 60° and 30" satd. (gross)	1100	960	850	1030	217

of part of the coke, and the Nielsen retort, where carbonisation is effected by the sensible heat of producer gas, are examples of the first case. In the "Pure Coal Briquette" process the rich gas which forms at the top of the vertical retort is diluted by higher temperature gas generated lower down the retort after the

[1] Burgess and Wheeler, Trans. Chem. Soc., 1910, 97, 1917-35.

[2] Maclaurin, R. "Low Temperature Carbonisation of Coal." Proc. S. Wales Inst. of Eng., 20th April, 1922.

greater part of the volatile matter has been removed by true low temperature distillation.

TABLE XXVII

COMPOSITION OF GASES COLLECTED IN VACUO BY BURGESS AND WHEELER[1]
DRY SILKSTONE COAL USED AND GAS CALCULATED AS NITROGEN FREE

Temperature ° C.	100	100 to 200	200 to 300	300 to 350	350 to 400
Gas :—					
Vol. @ N.T.P. per 100 grammes c.c.	34·0	65·5	58·5	985·0	4000·0
H_2S	—	—	—	1·70	0·70
CO_2	6·70	8·85	35·35	20·95	2·85
O_2	1·65	0·70	0·55	C_2H_2 0·15	C_2H_2 trace
C_2H_4	0·85	0·85	1·05	1·90	2·35
(n>2) C_nH_{2n}	1·30	2·90	18·85	· 17·90	6·15
CO	1·40	2·60	10·50	3·40	3·40
H_2	1·90	2·75	13·35	15·35	36·90
C_nH_{2n+2}	84·55	81·00	18·85	37·22	46·55
			——100·00——		
n/v for paraffins . (or number of CH_2 groups per unit volume of paraffins)	2·21	1·84	1·43	0·311	0·302

Appearance of H_2S was at about 270° C. and this is included in the figure for CO_2.

THE EFFECTS OF " CRACKING "

When the gas and tar vapour leaving the coal at a certain temperature is subjected for any length of time to the carbonising temperature, or to the effect of heat radiated from the sides of the retort, secondary changes take place which are generally classed under the name of " cracking." In these changes both gas and tar vapour are affected, and the general effect is the reduction of bodies of high molecular weight to others of lower molecular weight, with deposition of carbon and formation of lighter hydrocarbons and permanent gas. A similar effect is produced if the vapour when once formed is allowed to condense upon cooler coal

[1] Burgess and Wheeler. *Trans. Chem. Soc.*, 1914, **105**, 131–40.

and is later re-carbonised. The factors governing the changes which take place owing to the cracking of the rich gas and primary tar are many, and they are further complicated by the natural effect of increased temperature as already indicated.

In any carbonising system on a large scale, the formation of gas at different

TABLE XXVII

GASES COLLECTED BY BURGESS AND WHEELER[1]

COAL USED : ALTOFTS SILKSTONE SEAM

Temperature ° C.	450	500	600	700	750	800
Gas (N.T.P.) :— c.c. per gramme of ash free dry coal	13·0	29·9	99·00	124·00	154·00	218·00
NH_3	4·70	1·35	1·40	1·60	5·20	1·00
C_6H_6	8·60	4·85	5·20	3·40		3·65
CO_2	10·95	3·60	3·50	4·05	3·30	1·70
C_2H_2	Nil	0·35	—	0·40	—	—
C_2H_4	0·85	1·65	1·80	1·05	0·75	0·90
CO	8·75	6·45	7·10	7·90	9·40	11·85
H_2	7·00	16·60	26·60	32·70	41·65	48·55
CH_4	25·00	37·55	35·20	34·60	29·90	26·10
C_2H_6	34·10	27·60	19·20	14·30	9·80	6·25
Decrease in weight of coal %	9·1	18·79	28·37	32·39	34·04	36·30
Weight of tar collected	4·29	9·05	13·66	14·08	16·20	13·50

temperatures proceeds as indicated in Table XXVII, its composition being modified later by the cracking of the heavier hydrocarbons of both gas and tar to an extent which is dependent upon the particular process employed. In practically all low temperature systems the carbonising temperature is such that the gas will always contain a certain amount of the higher saturated hydrocarbons, particularly ethane.

[1] Burgess and Wheeler, Trans. Chem. Soc., 1910, 97, 1917-35.

THE EFFECTS OF STEAMING

Special tests which have recently been carried out by the Fuel Research Board, and which are described in Technical Paper No. 7, give interesting results of the analyses of the average gases produced from the carbonisation of coal at low temperatures in Glover-West vertical retorts with varying amounts of steaming.

TABLE XXIX

YIELDS OF GAS FROM LOW TEMPERATURE CARBONISATION IN GLOVER-WEST VERTICAL RETORT

Test No. .	1	2	3	4
Steam per cent .	Nil	7·24	13·47	20·00
Gas made per ton of coal, cu. ft. .	7190	6700	7350	7750
Gas made per ton of coal, therms	46·0	45·0	48·5	49·6
Gas made, calorific value : –				
B.Th.U.'s cu. ft. – gross .	640·0	671·0	661·0	640·0
nett .	585·0	602·0	594·0	573·0
Gas in therms as % of therms in coal .	16·5	16·0	17·2	17·7
Analysis.				
CO_2 .	4·9	5·6	5·6	7·2
C_nH_m	4·7	5·0	4·8	4·5
O_2 .	0·2	0·2	0·2	0·2
CO .	11·0	8·5	9·7	10·0
H_2 .	32·2	35·5	37·6	38·0
C_nH_{2n+2}	32·0	33·4	33·0	32·5
N_2 .	13·8	11·8	9·1	7·6
			100·0	
" n " in satd. hydrocarbons	1·09	—	—	—
Specific gravity (air=1)	0·572	0·544	0·561	0·563

The coal used was a mixture of 60 per cent Mitchell Main (caking) with 40 per cent Ellistown Main (non-caking), which had been proved to give satisfactory coke cakes on carbonisation at 600° C. in the horizontal retorts. The conditions of working in the vertical retorts were determined by a series of preliminary tests which indicated the desirability of maintaining a temperature of about 850° C. in the combustion chamber near the top of the retort, decreasing to about 700° C. at the bottom. Cokes applicable to household use were yielded in all cases. The yields

in each test are set out in Table XXIX, together with the analyses of the average gases.[1] The steam employed varied from nil to 20 per cent, the gas made ranging from 671 B.Th.U. per cubic foot with 7·24 per cent steam to 640 B.Th.U. with 20 per cent steam. The corresponding figure for no steam should have been 700 B.Th.U., which was actually obtained for a time, but a leakage of air into the retorts through the coke chambers reduced the average figure to 640 B.Th.U. only, as given in the table.

It will be seen by comparison with Table XXVI that the gases obtained from these experiments differ from normal low temperature gas in their much lower percentage of unsaturated hydrocarbons, the percentage being less than 5 per cent as compared with more than 9 per cent for H.M. Fuel Research Station horizontal retorts. The hydrogen content, on the other hand, is about doubled, being increased from some 17 per cent to an average of nearly 36 per cent. The amount of saturated hydrocarbons is much decreased—33 per cent as against 56 per cent—and those produced are of a lower mean molecular weight.

The results indicate that a setting of similar construction, but with iron retorts in which heat would be transferred to the coal much more rapidly, would possibly be suitable for the manufacture of good smokeless fuels by carbonisation of coal at low temperatures, provided the material to be carbonised can be delivered to the retort in such a form as will preserve an open charge which will not tend to fuse together into an unmanageable mass.

The volume of gas obtainable commercially of course varies according to the amount of cracking of the primary products which has taken place. We have already stated that the quality of the gas produced in the experimental setting of horizontal retorts at H.M. Fuel Research Station approaches more closely to that obtained in the laboratory than do that given in any other plant upon which independent tests have been carried out. The amounts yielded in this setting vary from 2700 cubic feet of gas per ton of coal, the gas having a calorific value of 1020 B.Th.U. per cubic foot or 27·54 therms of gas per ton, to 4220 cubic feet of gas per ton of coal, the gas having a calorific value of 906 B.Th.U. or 38·23 therms of gas per ton. These figures are typical of the gas obtainable by carbonisation of a wide range of coals at 600° C., and the enhanced figures for the therms of gas which are sometimes quoted should be accepted with great caution.

It is pointed out on page 28 that the rich gas obtained from a low temperature process has properties which make it exceptionally attractive as an enriching agent for water gas or for some of the comparatively low grade gases produced in certain systems of high temperature carbonisation.

[1] Fuel Research Board. Technical Paper No. 7. "Preliminary Experiments in the Low Temperature Carbonisation of Coal in Vertical Retorts." (H.M. Stationery Office.)

MOTOR SPIRIT FROM GAS

In all rich low temperature gas a certain quantity of the hydrocarbons is condensable by oil stripping at atmospheric temperatures, and after refining the resulting liquid is suitable for the production of motor spirit. The yields claimed by different systems are widely variant, but those given by the Fuel Research Board —1·43 gallons per ton of coal carbonised—may be quoted as an example which is backed up by experimental data. The question of the production of this liquid, however, and its characteristics, are described more fully in a later chapter.

THE PRODUCTS OF LOW TEMPERATURE CARBONISATION

TAR

INTRODUCTORY

As stated in the discussion of low temperature gas, the composition of the tar formed in any large-scale system depends to a certain extent on the conditions of that system, and particularly on the rapidity with which the tar is removed from the influence of heat and from secondary decomposition in the retort.

Accordingly, the best way to consider the composition of such tar is to examine the results obtained on an experimental scale where the conditions are more stringently defined, and where the effect of increasing temperature can be more carefully observed than in a plant under commercial or semi-commercial conditions. In this case all possible precautions can be taken, by vacuum distillation and otherwise, to remove the tar vapour from the retort before secondary decomposition has set in.

RESULTS OF INVESTIGATION ON LABORATORY-FORMED TARS

In 1904 Börnstein[1] fractionated eight Westphalian coals in 50° stages up to 450° C. and gave an account of the various tars collected. These, he stated, all contained paraffins and the higher phenols, but neither naphthalene nor anthracene.

In 1913 Pictet and Bouvier[2] examined a French bituminous coal by distillation in vacuo up to 430° C.

The thin tar collected amounted to 4 per cent by weight of coal. This tar was stated to consist chiefly of saturated and unsaturated hydrocarbons. Some oxygenated compounds were shown to be present, but no phenols.

Careful examination of the saturated hydrocarbons was later carried out by preliminary fractionation, and certain definite hydrocarbons were identified, such as hexahydromesitylene (C_9H_{18}) in the naphthene group, dihydrotrimethylbenzene (C_9H_{14}) in the unsaturated hydrocarbons, together with several paraffins. The similarity of the naphthenes to those present in American petroleum is pointed out.

An interesting experiment was also carried out in which the vacuum tar was cracked to give gas and a secondary tar. This secondary tar contained aromatic hydrocarbons, naphthalene, etc., as in normal gas tar prepared by high temperature distillation.

In 1914, this work was carried out on British coals by Wheeler[3] and others,

[1] Börnstein. Ber. d. Chem. Gesell., 913, **46**, 3342–53.
[2] Pictet and Bouvier. Compt. rend., Acad. Sci., 1913, **157**, 438–9.
[3] Burgess and Wheeler. "The Distillation of Coal in a Vacuum." Trans. Chem. Soc., 1914, **105**, 131–40

distillation in vacuo being again resorted to, and the temperature raised in 50° stages up to 450° C. In this case, however, the time of distillation occupied about five weeks, as compared with the five hours of Pictet and Bouvier. The yield of tar amounted to about 6·5 per cent by weight of coal, and this when fractionated gave the following results :—

(1) Unsaturated hydrocarbons of ethylenic character, but richer in carbon than C_nH_{2n}, amounting to 19-22 per cent of the yield.

(2) Naphthenes and liquid paraffins amounting to about 20 per cent of the yield.

(3) Tar acids, chiefly cresols and xylenols, amounting to about 7 per cent of the yield.

(4) Aromatic compounds which, however, did not contain naphthalene or anthracene, amounting to about 3 per cent of the yield.

(5) Solid paraffins of composition about $C_{26}H_{54}$ and M. Pt. 52·4 to 54° C. in small quantity.

(6) Pyridine bases—a trace.

(7) Pitch, boiling above 300° C. and completely soluble in chloroform, amounting to about 50 per cent of the yield. Sp. gr. at 15° C.=1·128.

The differences between this tar and that of Pictet and Bouvier seem to have been due either to the nature of the coal or to the time of distillation. These differences are chiefly in the absence of solid paraffins and phenols in the latter tar, but, as already stated, Börnstein also found these compounds in his distillations. This is particularly interesting in that Jones and Wheeler,[1] in 1915, obtained quite different tars by the separate distillation of " cellulosic " and " resinic " constituents of a coal. The " cellulosic " constituent yielded only very little tar, but this was composed chiefly of phenols. On the other hand, the yield of tar from the " resinic " bodies up to a temperature of 400° C. amounted to 40 per cent of the original weight, and was found to consist of paraffin, olefines and naphthenes, but no phenols.

In America, the chief workers in this field are Parr and Olin,[2] who first attacked the problem in 1907, through their researches on the manufacture of good smokeless fuel.

In their first paper they describe the form of retort employed and their method of carbonisation with superheated steam at 450° C., and give details of the yields, etc., obtained.

In a later paper the authors describe the manufacture of a good semi-coke by means of a similar process, and give a fuller description of the products obtained

[1] Jones and Wheeler. " The Composition of Coal." *Trans. Chem. Soc.*, 1915, 107, 1318–24.
[2] Parr and Olin. " The Coking of Coal at Low Temperature." *Bull.* 60 and 97, Univ. of Illinois.

at 400–500° C. Thus they obtained from Illinois bituminous coal a dark brown tar of 1·069 sp. gr$_t$, which they separated primarily into three fractions as follows :—

	Percentage of Tar.	Sp. Gr.
Light oil to 210° C.	17·2	0·966
Heavy oil to 210–325° C.	52·7	1·032
Pitch over 325° C.	30·1	1·270

From this tar the yields obtained by refining and separation were as follows :—

		Percentage of Tar.
	Phenols, etc.	5·7
	Amines and bases	0·9
	Fraction 75– 95° C.	0·38
Light oil	95–125° C.	1·33
	125–170° C.	4·77
	170–200° C.	2·67
	200–210° C.	1·32
	Tar acids	22·2
Heavy oil	Fraction 210–250° C.	2·87
	250–270° C.	13·55
	over 270° C.	1·53

The authors report the presence of pentane and hexane as well as benzene in the fraction 75–95° C. and of heptane in the fraction 95–125° C. The two following fractions are stated to consist of benzene, toluene and the xylenes. The tar acids were found to contain a high proportion of cresols. Naphthalene and anthracene were absent.

The presence of benzene and toluene in quantity in this tar is remarkable in that the carbonising temperature was not greater than 500° C., but may have been due to overheating of the tar vapours in the retort. In this connexion Whitaker and Crowell[1] state from their investigation of a Pennsylvanian coal that these hydrocarbons are first formed at 500° C. and 400° C. respectively, and that maximum yields are obtained by carbonisation at 800° C. for benzene, 700° C. for toluene and 600° C. for xylenes. If this be so, true low temperature tar may contain appreciable quantities of all three products.

In Germany, the chief workers have been Fischer and Gluud,[2] and they have directed special attention to the possible yields of light hydrocarbons. The following

[1] Whitaker and Crowell. *J. Ind. Eng. Chem.*, 1917, **9**, 261–3.
[2] Fischer and Gluud. *Ber.*, 1919, **52**, 1053, 1068 ; *Ges. Abh. z. Kennt. d. Kohle*, 1918, **3**, 46.

yields were obtained with three different coals, the distillation range of the liquid being such that 95 per cent boiled below 190° C. :—

	Yield, percentage by weight of coal used.
Gas coal (Lohberg Mine) .	1·23
Bituminous coal (Osterfeld Mine)	0·67
,, ,, (Minden Mine)	1·00

These " spirits " on fractionation yielded the following :—

20–60° C.	. . .	Paraffins, chiefly C_5H_{12} and C_6H_{14} and some unsaturated hydrocarbons. Density about 0·65 at 8° C.
60–100° C.	. . .	Mixture of paraffins, naphthenes and some nitrogen and sulphur compounds. No benzene was found. Density about 0·72 at 20° C.
100–125° C. 125–190° C.	. . .	Paraffins, naphthenes and complex aromatic hydrocarbons. Density about 0·77 and 0·87 respectively at 20° C.

The authors also describe certain properties of the higher fractions obtained from tar and discuss the separation of certain solid paraffins ranging from $C_{24}H_{50}$ to $C_{29}H_{60}$, which amounted to 0·4 per cent of the bituminous coal tar and to 1·5 per cent of the gas coal tar. It should be noted that again no naphthalene or anthracene was detected.

NITROGENOUS AND SULPHUR BODIES

As stated by Jones and Wheeler,[1] the percentage of nitrogenous bodies in low temperature tars is less than one. This has been borne out by the work of other investigators, as shown by the following list :—

	Per cent.	
Parr and Olin	0·9	Amines
Pictet, Kaiser and Labouchère	0·2	Bases
Gluud	0·46	Pyridine bases
Morgan and Soule	0·32	Bases

[1] Jones and Wheeler. "The Composition of Coal." *Journ. Chem. Soc.*, 1914, **105**, 141.
[2] Parr and Olin. "The Coking of Coal at Low Temperature." *Bulletin* 60, Univ. of Illinois.
[3] Pictet, Kaiser and Labouchère. *Compt. rend.*, 1917, **165**, 113.
[4] Gluud. *Ges. Abh. z. Kennt' d. Kohle*, 1918, **3**, 46.
[5] Morgan and Soule. "Examination of Low Temperature Coal Tars." *J. Ind. Eng. Chem.*; 1923, **15**, 587–91.

The most complete work on the isolation of these bodies from tar is that of Pictet and his co-workers. They found no pyridine, but isolated a number of saturated hydro-derivatives of quinoline and iso-quinoline.

No literature is at present available on the sulphur bodies present, but the amount of these is very small.

CHARACTERISTICS OF LOW-TEMPERATURE TAR OBTAINED FROM A LARGE-SCALE PLANT

From what has already been said, it is obvious that the nature of the low temperature tar produced on a large scale will depend to some extent on the rapidity with which the vapours are removed from the retorts, and therefore from the possibility of secondary decomposition.

In any tar prepared at low temperatures the following general characteristics may, however, be expected :—

(1) That it will contain hydrocarbons chiefly of the paraffin and olefine series with some naphthenes and from 10–15 per cent of phenols.

(2) That the quantity of benzene and its homologues will be small, but will be made up for by the presence of light paraffins such as hexane, etc.

(3) That the higher phenols, such as cresol, will be more represented than phenol itself, and there will also be present certain polyhydric alcohols.

(4) That the content of free carbon will be less than 2 per cent, and that of pitch will be 35–40 per cent. ·

(5) That there will be up to 1 per cent of basic nitrogenous substances including pyridine.

If, however, the tar has been subjected to overheating in the state of vapour or to condensation and re-distillation in the retort, it will contain small quantities of the substances more particularly identified with high temperature tar. These are the aromatic hydrocarbons, naphthalene, anthracene, etc. It will also contain an increased amount of pitch and free carbon.

In many of the present commercial scale systems the tar has been treated entirely as a by-product, and attention directed chiefly to the production of good coke. It is therefore only in certain processes that particulars are available of the tars produced, and these are set out for comparison in Table XXX

A more complete examination of the tar produced from several English coals in semi-large scale work at 600° C. has been carried out at H.M. Fuel Research Station.[1] The tars produced a e typical of straight low temperature carbonisation,

[1] Fuel Research Board. Report for Years 1920, 1921, Second Section : "Low Temperature Carbonisation." (H.M. Stationery Office.)

and the yields and nature of the tars, together with the details of refinery and distillation of the fractions, are given in Tables XXXI–XXXV.

From these results, only a slight idea can be formed as to whether or not the tars produced on a larger scale are identical with those already described in the work of different investigators on laboratory-formed tars. So far as the results go, the systems in which the products of carbonisation are removed as rapidly as possible produce tars which are quite comparable with those quoted from the Fuel Research Board report.

TABLE XXX

MAIN DETAILS OF TARS FROM CERTAIN FULL-SCALE PROCESSES

Process.	H.M. Fuel Research Station.		Tozer System.		"Sensible Heat" (Nielsen) System.		Carbocoal.
Sp. Gr. @ 60° F.	1·033		1·060		1·076		...
Calorific Value, B.Th.U.'s per lb.	16,840		—		16,000		——
Distillation :	%	Sp. Gr.	%	Sp. Gr.	%	Sp. Gr.	%
0–170° C.	9·1	0·842	6·0	0·80	4·75	0·80	4·7
170–230° C.	19·4	0·969	16·5	0·97	20·0	0·87	13·7
230–270° C.	12·8	0·998	9·0	0·97	14·5	0·97	11·6
270° C.–Pitch	11·8	1·027	36·0	1·04	24·0	1·01	27·0
Pitch	46·6	1·197	32·5	—	35·0	—	43·0
Loss	0·3	—	—	—	1·75	—	——
Tar Acids per cent of Tar	17·3		10·22		20·0		35% of fraction 170–360
Motor Spirit (crude) :							
From gas, galls. per ton	1·68		2·6		—		} 2–3
From tar, ,, ,,	1·27		1·3		—		

TABLE XXXI

ANALYSES OF TAR FROM TESTS IN HORIZONTAL RETORTS

	Langley Brights.		Rossington Main.		Dalton Main 1.		Dalton Main 1.		Dalton Main 2.		Mitchell Main.		Ellistown Main.		60 per cent Mitchell Main. 40 per cent Ellistown Main.	
	% by wt.	Sp. gr. at 15° C.	% by wt.	Sp. gr. at 15° C.	% by wt.	Sp. gr. at 15° C.	% by wt.	Sp. gr. at 15° C.	% by wt.	Sp. gr. at 15° C.	% by wt.	Sp. gr. at 15° C.	% by wt.	Sp. gr. at 15° C.	% by wt.	Sp. gr. at 15° C.
Temperature	580° C.		600° C.		550° C.		600° C.		600° C.		600° C.		600° C.		600° C.	
Tar (dry):— Specific gravity at 15° C.	1·030		1·031		1·020		1·030		1·033		1·036		1·053		1·057	
Cal. value B.Th.U. per lb.	16,600		15,200		16,650		16,570		16,840		16,590		16,020		16,360	
Sulphur per cent	0·61		0·62		0·45		0·50		0·75		0·43		0·36		0·73	
Distillation:— To 170° C.	3·3	—	7·4	—	16·6	0·854	13·2	0·840	9·1	0·842	10·6	0·840	7·6	0·851	8·6	0·842
170–230° C.	19·5	—	22·6	0·930	13·7	0·975	15·1	0·972	19·4	0·969	16·1	0·958	18·1	0·980	16·3	0·966
230–270° C.	15·3	—	12·9	1·022	14·1	0·995	10·7	0·997	12·8	0·998	13·9	0·990	13·8	1·004	13·5	0·997
270–350° C.	—	—	—	—	—	—	—	—	11·8	1·027	12·3	1·021	18·1	1·036	15·0	1·032
270–350° C.	25·0	—	9·0	—	20·6	1·003	22·6	1·005								
Pitch	30·1	—	42·4	—	30·1	—	30·2	—	46·6	1·197	45·1	—	41·6	—	45·7	1·212
Loss	6·8	—	5·7	—	5·0	—	8·2	—	0·3	—	2·0	—	0·8	—	0·9	—
	100·0		100·0		100·0		100·0		100·0		100·0		100·0		100·0	

TABLE XXXII°

PURIFICATION OF FRACTIONS FROM DISTILLATION OF LOW TEMPERATURE TARS

Coal.		Loss on extraction as percentages by volume of original fractions.			
		Fraction			
		1. To 170° C.	2. 170-230° C.	3. 230-270° C.	4. 270-310° C.
Dalton Main 2 . . .	Caustic soda . .	4·0	43·0	40·5	26·0
	Sulphuric acid . .	7·0	3·0	5·5	6·0
	Washed oil remaining ,	89·0	54·0	54·0	68·0
	Washed and refined oil	83·0	54·0	51·5	66·1
Mitchell Main . . .	Caustic soda . .	2·5	43·5	39·0	26·5
	Sulphuric acid . .	11·5	2·6	5·5	5·5
	Washed oil remaining .	86·0	53·9	55·5	68·0
	Washed and refined oil ·	76·5	51·4	52·2	61·4
Ellistown Main . .	Caustic soda . .	7·3	52·0	50·1	36·0
	Sulphuric acid . .	9·7	4·0	5·4	6·0
	Washed oil remaining.	83·0	44·0	44·5	58·0
	Washed and refined oil	70·8	38·7	40·3	51·4
60 per cent Mitchell Main . 40 per cent Ellistown Main	Caustic soda . .	4·3	46·0	44·0	30·5
	Sulphuric acid . .	6·8	3·5	5·0	6·5
	Washed oil remaining .	88·9	50·5	51·0	63·0
	Washed and refined oil	76·5	46·4	45·6	57·7

THE USES OF LOW TEMPERATURE TAR

(q) DIRECTLY AS FUEL

Low temperature tar as made is suitable as a fuel for Diesel engines once it has been dehydrated and freed from suspended solid matter. Its calorific value ranges from 16,000 to 13,500 B.Th.U per lb., but owing to its flash point being below the atmospheric temperature it does not conform to the Admiralty specification of 175° F. Any attempt to remove the light spirit from the tar to correct

TABLE XXXIII

Low TEMPERATURE TAR. DISTILLATION RANGES OF WASHED OILS FROM CRUDE FRACTIONS

Temperatures °C.	Dalton Main 2.	Mitchell Main.	Ellistown Main.	Mixture of Mitchell Main and Ellistown Main.
1st Fraction (to 170° C.)				
---	95° C.	87° C.	92° C.	90° C.
	1st drop	1st drop	1st drop	1st drop
100	0·5	1·0	0·8	1·0
110	2·0	3·4	2·9	3·2
120	22·3	24·9	12·9	17·1
130	53·6	55·0	42·1	45·1
140	79·4	76·5	66·6	70·9
150	93·0	90·2	84·1	87·9
160	98·5	96·5	94·2	96·1
---	---	98·9	98·0	98·5
---	161° C.	166° C.	168° C.	167° C.
---	Flask dry	Flask dry	Flask dry	Flask dry
Specific gravity at 15° C.	0·829	0·823	0·831	0·825
2nd Fraction (170–230° C.)				
170	1st drop	2·4	1st drop	1·0
180	1·0	9·0	2·0	4·8
190	10·9	28·5	11·9	23·2
200	30·6	45·6	28·4	42·3
210	42·6	59·3	42·9	54·1
220	52·6	69·8	54·9	64·9
230	61·5	77·7	65·5	7·8
270	83·0	93·9	88·1	92·0
300	90·0	---	---	---
Specific gravity at 15° C.	---	0·893	0·898	0·894

(Continued on next page.)

TABLE XXXIII (*continued*)

Temperatures °C.	Dalton Main 2.	Mitchell Main.	Ellistown Main.	Mixture of Mitchell Main and Ellistown Main.
3rd Fraction (230–270° C.)				
220	2·9	10·8	5·9	4·0
230	10·3	23·9	13·5	12·5
240	22·5	41·3	26·6	27·6
250	41·2	56·0	41·7	43·0
260	55·0	68·9	55·7	51·7
270	67·8	78·0	66·3	67·8
280	76·5	84·6	75·4	76·8
290	81·5	89·0	81·8	81·7
300	86·4	92·0	86·3	85·8
310	—	94·1	90·6	89·3
Specific gravity at 15° C.	—	0·954	0·955	0·953
4th Fraction (270–310° C.)				
230	—	—	3·0	—
240	—	2·8	—	—
250	2·3	7·9	6·0	2·8
260	5·3	15·6	8·0	7·0
270	15·2	26·9	12·9	15·2
280	28·4	42·0	21·6	28·4
290	42·5	55·4	32·5	40·4
300	56·2	66·1	44·4	53·0
360	93·6	92·2	88·5	91·5
Specific gravity at 15° C.	—	0·992	0 991	0·995

TABLE XXXIV

LOW TEMPERATURE TAR. DISTILLATION RANGES OF SEPARATE TAR ACIDS
(PERCENTAGE BY VOLUME)

Distillation temperatures.	Dalton Main 2.	Mitchell Main.	Ellistown Main.	60 per cent Mitchell Main. 40 per cent Ellistown Main.
From crude fraction 170–230° C.				
0–191° C.	6·9	14·1	8·7	3·5
0–196	16·8	34·7	14·4	5·9
0–198	29·5	47·0	23·4	13·2
0–200	43·7	56·8	37·2	25·7
0–202	52·2	64·0	47·2	36·0
0–204	61·1	70·0	55·6	50·3
0–210	—	82·5	73·7	71·9
0–220	—	90·4	87·3	86·2
From crude fraction 230–270° C.				
0–210° C.	—	34·2	8·4	15·6
0–220	—	62·3	43·8	50·9
0–230	—	78·2	63·7	69·6
0–240	—	83·2	73·8	79·4
0–250	—	87·7	79·4	84·5
0–260	—	89·5	83·7	87·6
0–270	—	91·6	86·6	89·4
From crude fraction 270–310° C.				
0–230° C.	—	5·1	4·7	3·6
0–270	—	35·6	26·6	35·3
0–310	—	62·6	63·5	68·0

SPECIAL FRACTIONATION OF TAR ACIDS FROM DALTON MAIN Coke 2

	Fraction.		
	To 204° C.	204–230° C.	230–310° C.
Percentage by volume of tar	5·43	5·94	4·97
Gallons per ton of dry coal	0·71	0·77	0·64

this would result in so much thickening that the residue would fail to pass the tests for viscosity.

The adaptability of the crude oil for mixture with natural oils has been investigated at H.M. Fuel Research Station, and it has been there found that out of seven oils chosen, two (American and shale oils) were quite immiscible, while with Burmese, Texas and Mexican oils the only stable mixtures were those containing less than 25 per cent of crude oil. With Persian oil up to 10 per cent of tar can be added, but separation occurs in all other mixtures. With only one oil—Trinidad—was complete miscibility found, and even in this case the application of heat was necessary. Details of these miscibility tests are given in Table XXXVI.[1]

TABLE XXXV

WASHED AND REFINED OILS FROM TAR CALCULATED TO GALLONS PER TON OF DRY COAL

Coal carbonised.	Dalton Main 2.	Mitchell Main.	Ellistown Main.	60 per cent Mitchell Main 40 per cent Ellistown Main.
Original yield of tar . . .	14·00	16·25	12·87	15·51
Fraction 0–170° *C*				
Original fraction . . .	1·56	2·13	1·22	1·68
Washed spirit . . .	1·39	1·83	1·01	1·49
Washed and refined spirit . .	1·30	1·63	0·86	1·29
Fraction 170–230° *C.*				
Original fraction. . .	2·90	2·83	2·53	2·78
Washed oil . . .	1·57	1·53	1·11	1·40
Washed and refined oil . .	1·57	1·46	0·98	1·29
Fraction 230–270° *C.*				
Original fraction . . .	1·85	2·36	1·88	2·24
Washed oil . . .	1·00	1·31	0·84	1·14
Washed and refined oil . .	0·96	1·23	0·76	1·02

[1] Fuel Research Board. Report for Years 1920, 1921; Second Section : "Low Temperature Carbonisation." (H.M. Stationery Office.)

Coal carbonised.	Dalton Main 2.	Mitchell Main.	Ellistown Main.	60 per cent Mitchell Main. 40 per cent Ellistown Main.
Fraction 270–310° *C.*				
Original fraction	1·66	2·63	2·39	2·39
Washed oil	1·13	1·38	1·39	1·51
Washed and refined oil	1·10	1·25	1·23	1·38
Total of original fractions	7·97	9·35	8·02	9·09
Total washed and refined oils	4·93	5·57	3·83	4·98
Total tar acids crude	2·49	2·74	3·20	3·07
Total tar acids refined	2·12	2·34	2·58	2·61
Pitch, lb. per ton of dry coal	67·5	75·9	56·9	74·9

SPECIALLY SEPARATED OILS FROM DALTON MAIN TAR 2

	Sp. gr. 15° C.	Per cent by volume of tar.	Galls. per ton of dry coal.
Light naphtha to 160° C.	0·829	8·25	1·15
Heavy naphtha, 160–200° C.	0·870	4·59	0·64
Burning oils, 200–270° C.	0·936	11·93	1·67
Gas oil, 270–300° C.	0·979	5·46	0·76
Light lubricating oil, 300–360° C.	1·005	5·49	0·71
Total		35·33	4·93

TABLE XXXVI

MISCIBILITY OF LOW TEMPERATURE TAR WITH NATURAL FUEL OILS

Name.	Nature.	Fuel -Oil. Sp. gr. 15° C.	Proportions mixed.		Separation.	
			Tar.	Oil.	Amt.	Nature of.
Shale	Very fluid	0·867	10	90	6	Semi-solid.
			25	75	11	,, ,,
			50	80	14	Black thick fluid.
			70	30	5	,, ,, ,,
American	Very fluid	0·902	10	90	9	Resinous and gummy.
			50	50	19	,, ,, ,,
			65	35	18	Thick and gummy.
			75	25	4	Black and fluid.
Burmah	Very fluid	0·895	10	90	8	Gummy, resinous.
			25	75	12	Semi-solid.
			50	50	18	Thick, but fluid.
			65	35	24	,, ,,
			75	24	—	No separation.
Texas	Fluid	0·926	10	90	11	Thick and gummy.
			25	75	12	,, ,, ,,
			50	50	20	Thick, fluid.
			65	35	18	,, ,,
			75	25	—	No separation.
Mexican	Very thick	0·936	10	90	5	Thick, but fluid.
			25	75	10	Fluid.
			50	50	—	No separation.
			65	35	—	,, ,,
Persian	Thick	0·942	10	90	—	No separation.
			25	75	10	Thick and fluid.
			50	50	20	,, ,,
			75	25	25	,, ,,
Trinidad	Very thick	0·968	10	90	—	No separation, when heated during mixing
			50	50	—	,, ,, ,,
			75	25	—	,, ,, ,,

Although the crude oil from low temperature carbonisation is not usually miscible with natural petroleum, several methods of treatment have been from time to time suggested in order to overcome this difficulty. In the Lessing[1] process, tar is mixed with a petroleum spirit which causes separation of the pitch, the mixture of oils and solvent being then subjected to steam distillation for recovery of the latter.

A sample of low temperature tar obtained from Arley coal was treated by Dr. Lessing according to the above process. This process consists in separating oils and pitch from tar by treatment with petroleum spirit free from aromatic hydrocarbons, whereby the pitch is precipitated and all the oils are dissolved. The process is carried out at a temperature above the melting point of pitch and below the initial boiling point of the solvent, and thus a sharp separation of pitch and oils is obtained at a temperature of approximately 100° C. Tar acids and bases may be extracted from the oil solution and the solvent subsequently distilled off, condensed and re-used without cooling, all operations being carried on in a continuous cycle. The tar acids and bases may alternatively be extracted after the removal of solvent.

The following products were obtained :—

(1) Liquor.
(2) Crude Oil, i.e. oil after separation of pitch and low boiling fractions to 140° C.
(3) Neutral Oil, i.e. crude oil after removal of tar acids and bases.
(4) Tar Acids.
(5) Tar Bases.
(6) Pitch.

The yields were as follows :—

Crude tar oil 80·7% { Neutral oil 56·1% / Tar acids 24·4% / Tar bases 0·2% }

Pitch 16·9%

By ordinary distillation this tar yielded 44·2 per cent by weight of pitch. When the oils are distilled from an Engler flask only about 70 per cent can be distilled below 360°, leaving 30 per cent pitch. This, with the 16·9 per cent removed in the Lessing process, corresponds to 43·1 per cent on the original tar.

The crude oil and neutral oil were both found to be miscible in all proportions with a typical petroleum fuel oil. The oil solution was " cut " so as to minimise the loss of the low boiling constituents of the original tar. Consequently no attempt was made to bring the oil within the Admiralty specification as regards flash-point. By fixing a " cutting " point, which will make the oil comply with the specification,

[1] Lessing, R. Patent No. 130,362.

M

a certain percentage of solvent is furnished by the lower boiling constituents of the tar itself.

The viscosity and setting points of both crude and neutral oils are well within the Admiralty specification.

It is as yet too early to form any accurate idea as to whether the cost of treatment by this method would be such as to make it commercially practicable, but experimental work is going on at H.M. Fuel Research Station and, elsewhere, upon the lines indicated.

(b) As Fuels after Distillation

It is not necessary, however that the tar should be used in the crude state. By fractionation and refining good fuel and lubricating oils can be obtained, as well as a quantity of light spirit which, when added to that obtainable by scrubbing the gas, gives a mixture whose properties are very similar to those of a good quality petrol, and so is very suitable for use as motor spirit. The figures already shown in Table XXXV and presented in slightly modified form in Table XXXVII are those obtainable by fractionation and refining of a tar which was obtained by the carbonisation of Dalton Main coal. The losses by refining are of course considerable, but the products are of good quality.

TABLE XXXVII

Separated Products from Tar from Dalton Main Coal

Total tar per ton 14 galls.	Sp. gr. 15° C.	Galls. per ton of Dry Coal.
Light naphtha, to 160° C.	0·829	1·15
Heavy naphtha, 160–200° C.	0·870	0·64
Burning oils, 200–270° C.	0·936	1·67
Gas oil, 270–300° C.	0·979	0·76
Light lubricating oil, 300–360° C.	1·005	0·71
Total		4·93
Loss in refining	—	3·41
Tar acids	—	2·12
Total		10·46
Pitch	—	67·5 lb.

(c) Tar Acids

The tar acids amounting as they do to 15 to 25 per cent of the total tar made, may also be found to have a considerable commercial application, as, for example,

in the manufacture of disinfectants. The actual yields of tar acids quoted in the literature available on the subject seem to vary rather widely. Some of these figures are shown in Table XXXVIII.

TABLE XXXVIII

YIELDS OF TAR ACIDS FROM LOW TEMPERATURE TAR

	Tar Acids as per cent of Tar.
Jones and Wheeler (Vacuum Tar)	6–7
Parr and Olin	27·9
S. R. Church, Illinois	50·0
Carbocoal process	14·7
Tozer process	22·0
" Sensible Heat " (Nielsen) process . .	20·0
Fuel Research Station	15–25

In view of the wide variation in the yields, the figures accepted as probably representing average values are those of H.M. Fuel Research Station. The highest figure was obtained with a coal of high oxygen content. It is possible, therefore, to obtain by low temperature carbonisation a yield of valuable tar acids up to 3 gallons per ton of dry coal carbonised.

The composition of these phenols has not been determined on any tar made by a large-scale process, but several investigators have indicated the substances obtained on an experimental scale. Jones and Wheeler in vacuum tar found chiefly cresols and xylenols. Pictet found no lower phenols in his tar except after it had been standing for several years, when he found phenol and the three cresols.

Fischer and others have indicated the possibility of yields up to 50 per cent of the tar and have made a rough separation of the compounds as follows :—

	Per cent of Tar.
Cresols	1–2 (chiefly m-cresol)
Xylenols . . .	1–2
Higher Phenols . . .	30–32 (containing trimethylphenols)
Acid Resins . .	10

Morgan and Soule give the following composition of phenols found in tar from the Carbocoal process :—

	Per cent of Tar.
Phenol . . .	0·6
Cresols . . .	4·9
Xylenols . . .	2·8
Higher Phenols .	5·1
Resins . .	1·3
	14·7

In this case the lower homologues are in much higher proportion, and also all three cresols were in evidence in the proportions of 27 per cent ortho, 19 per cent meta, and 54 per cent para, or in contradiction to Fischer's tar, with the m-cresol in the minority. In the tars examined at the Fuel Research Station no phenol was present, about 5 per cent of cresols and 11 per cent of xylenols and higher phenols.

Low temperature tars have been shown to be particularly useful in the manufacture of disinfectants. It may probably be assumed that this is due to the high bacterial co-efficients of the higher phenols and the high proportion in which they are present.

YIELDS OF LOW TEMPERATURE TAR

The yields of tar obtained in H.M. Fuel Research Station horizontal setting from a variety of coals, vary from 13·7 gallons to about 15 gallons per ton of coal carbonised, being about 60 per cent of that given by similar coals carbonised in the laboratory assay apparatus. It is of course not impossible that, by modification of the plant, yields more nearly approaching the theoretical might be obtained ; but the figures quoted above illustrate the difficulty which is always experienced in large-scale plant in obtaining the maximum tar yields. In their estimates of the quantity of the products obtainable from low temperature carbonisation if and when it is established on a national scale, the Fuel Research Board adopt 14·5 gallons per ton of oil as the average quantity which could be relied upon from a fairly wide range of coals.

CHAPTER XIV

THE PRODUCTS OF LOW TEMPERATURE CARBONISATION

MOTOR SPIRIT

INTRODUCTORY

MOTOR spirit is available from both the gas and tar of low temperature carbonisation processes. From the latter its recovery is simple, and practically the whole of the tar fraction to 170° C. can, when rectified, be utilised. This fraction, as shown in Table XXXV, amounts to about 10 per cent of normal tar production ; the available yield of fine spirit is therefore of the order of 1·0-1·6 gallons per ton of coal carbonised. This refined spirit consists chiefly of the higher paraffins with some benzene and toluene and open chain unsaturated compounds.

RECOVERY OF MOTOR SPIRIT FROM GAS

The spirit in the gas can be recovered only by some scrubbing or condensation process. An example has already been quoted of the first procedure in which paraffin oil was used as the scrubbing or condensing agent, and the spirit obtained by rectification.[1] In this experimental work the yield of light spirit represented 0·67-1·23 per cent of the coal carbonised, or from 2·0-3·8 gallons per ton of coal. In composition the spirit proved to be a mixture of saturated and unsaturated hydrocarbons, naphthenes, and complex aromatic hydrocarbons. Benzene was not present in any of the spirits examined, although its presence in small quantities in the products of low temperature distillation is noted by Wheeler, Parr, Olin and others.

In the Report of the Fuel Research Board for 1920, 1921,[2] an account is given of the recovery of spirit by means of scrubbing with gas oil the low temperature gas made from Dalton Main coal. The work was carried out on a small scale, and gave a yield of refined spirit of 1·40 gallons per ton of coal at a specific gravity of 0·731 at 15° C. This work has since been repeated upon a larger scale, and a similar return obtained. The details of the small-scale test are shown in Table XXXIX[2] together with the distillation range of the refined spirit.

Other methods for the recovery of this light spirit from gas have been suggested, and in particular activated charcoal has been used by several workers as a condensing medium. A. Engelhardt[3] has described its use for the recovery of benzol from illuminating gas in a process whereby the gas is passed through activated

---

[1] Fischer and Gluud. "Meine Forschungsergebnisse auf dem Gebiet der Kohlenextraktion." *Glückauf*, **52**, 721-9.

[2] Fuel Research Board. Report for Years 1920, 1921; Second Section: "Low Temperature Carbonisation." (H.M. Stationery Office.)

[3] A. Engelhardt. "Benzene Recovery by means of Active Carbon.", *Gas Journal*, **159**, 421; *Gas u. Wasserfach*, 1922, **65**, 473.

charcoal, the condensed vapours being liberated after a suitable period by the action of superheated steam. After drying, the charcoal is used again. A graph is there given showing the sorptive power of charcoal for benzene, alcohol, etc. The

<div align="center">TABLE XXXIX</div>

<div align="center">SCRUBBING TESTS ON LOW TEMPERATURE COAL GAS FROM DALTON MAIN COAL 2</div>

<div align="center">Gas yield :— 3200 cu. ft. per ton of dry coal.</div>

	Test 1.	Test 2.
Duration of test, hours . . .	8·1	5·0
Gas :—		
Total amount scrubbed cu. ft. . .	49·7	60·0
Rate in cu. ft. per hour . . .	6·12	12·0
Wash Oil :—		
Rate in ccs. per hour . . .	1394	1608
Cc. per cu. ft. of gas . . .	229	134
Final concentration of spirit, per cent	1·04	1·60
Yield of Spirit :—		
Total yield in cc. of crude spirit .	118·6	130·8
,, ,, galls. per ton of coal .	1·68	1·54
Refined spirit in cc. . . .	102·0	118·0
,, ,, ,, galls. per ton . .	1·45	1·39
,, ,, Sp. gr. 15° C. . .	0·731	0·731
Engler distillation of Refined Spirit :—		
40° C. . . .	First drop	
To 60° C. . . :	8·8%	
,. 70° C. . . .	23·3%	
,, 80° C. . . .	42·0%	
,, 90° C. . . .	56·7%	
,, 100° C. . . .	68·0%	
,. 110° C. . . .	77·8%	
,, 120° C. . . .	85·0%	
,, 130° C. . . .	91·0%	
,, 140° C. . . .	94·3%	
Flask dry at 145° C. . . .	96·0%	

concentration of solvent vapour in normal low temperature coal gas is of the order of 60–80 grammes per cubic metre, and it may be seen from the graph that fairly high sorptive powers may be obtained for benzene at this concentration. Whether the hydrocarbons present in low temperature gas are as readily condensable yet remains to be determined. Should they prove to be so, the method would appear to be distinctly promising.

QUALITIES OF THE SPIRIT AS A FUEL FOR INTERNAL COMBUSTION ENGINES

No adequate work has yet been undertaken on the applicability of low temperature spirit for the purposes of internal combustion engines. Preliminary tests, however, which have been carried out at H.M. Fuel Research Station, indicate that it is likely to prove entirely suitable.

When the distillation ranges of the refined spirits are examined it is seen that the spirit collected from the gas is extremely light. So much is this the case that, when mixed with the heavier tar spirit, a mixture is produced which is similar in its main properties to the petrol at present marketed. The presence of an appreciable light fraction is, moreover, a decided advantage from the point of view of the starting of a cold engine. Table XL gives the results of a few experiments which indicate that the explosive range of the spirit is greater than that of petrol or benzol.

TABLE XL

EXPLOSIVE RANGE OF SOME MOTOR SPIRITS

Spirit	Explosive Range. c.c. Spirit per cu. ft. of Air at 15° C.
Benzol, Sp. Gr. 0·881 . .	2·7 to 6·1
Petrol B.P. No. 1, Sp. Gr. 0·723	2·5 to 7·3
Spirit from Gas . . .	2·3 to 9·5
Mixture 50/50 . . .	2·5 to 10·5

As regards miscibility with other motor fuels, the following conclusions have been formed :—

(1) That the spirit is miscible in all proportions with petrol and the benzol of the British Standard specification.

(2) That with alcohol the miscibility depends on the strength of the alcohol. The solubility of the spirit at —10° C. in alcohol is as follows :—

	Spirit from Gas.	Spirit from Tar.
Alcohol 95% by vol.	74·2	—
,, 90% by vol.	16·3	—
,, 85% by vol.	7·1	25·7

(3) That with benzol and 95 per cent alcohol mixed in equal proportions the miscibility of the spirit at —10° C. was 60 per cent, or the mixture contained :—

Spirit	.	.	.	.	60
Alcohol 95%	.	.	.	.	20
Benzol 90%	.	.	.	.	20

(4) That the slight tendency which even refined spirit has to darken on standing owing probably to resinification taking place, is prevented by the alcohol ; or, in other words, the resins formed are not deposited, but retained in solution by the alcohol.

It would appear, therefore, that, so far as experimental work has yet indicated, the spirit from low temperature carbonisation is suitable for use in internal combustion engines, either alone or in admixture with alcohol, benzol, or petrol.

The total yield of motor spirit from gas and tar under favourable conditions will amount to about 3 to 4 gallons per ton of coal carbonised.

NITROGENOUS PRODUCTS FROM COAL

INTRODUCTORY

THE ammonia obtained in most processes of low temperature carbonisation does not form an asset of any importance in the balance sheet; indeed at present prices it would in many cases hardly pay for its recovery. In a limited number of processes, however, the conditions are such as to render it possible to obtain yields approaching those of coke ovens or gas works, but even then it will often be found that disadvantages have presented themselves which more than offset the increase in value in the ammonia yield.

In order to study the subject of ammonia production by low temperature carbonisation, it is necessary to discuss the matter from a fairly wide point of view in order to see clearly the reasons which lie behind the low recovery of nitrogen in the subject under consideration. The present chapter will therefore be devoted to a short discussion of ammonia recovery in the various processes of gas and coke manufacture in addition to that of low temperature carbonisation.

THEORETICAL AMOUNT OF AMMONIUM SULPHATE FROM COAL

The usual nitrogen content of bituminous coals varies from about 1·3 per cent to 1·6 per cent, or from 29 lb. to 36 lb. per ton of coal. Were it possible to convert the whole of this into ammonia, the production would lie between 35 lb. and 44 lb. of ammonia, corresponding to ammonium sulphate yields of 137 lb. and 170 lb. per ton of coal.

YIELD OF AMMONIUM SULPHATE IN THE CARBONISATION INDUSTRIES

In the ordinary carbonisation industries, the production of ammonia, which is shown in Table XLI from the Final Report of the Nitrogen Products Committee, 1920, falls far below that theoretically possible from the nitrogen in the coal, and

TABLE XLI

PRODUCTION OF AMMONIUM SULPHATE FROM COAL
(Nitrogen Products Committee)

Industry.	Year.	Ammonium Sulphate (lb. per ton of Coal).
Gas Works	1911–13	22·7–23·0
,, ,,	1917	22·4
Coke Ovens (All types)	1917	18·0
,, ,, (Recovery)	1917	24·0
Gas Producers	1911–13	·84·0
,, ,,	1917	66·3

for the usual gas works and coke oven practice amounts to little more than 20 lb. per ton of coal. For gas producers, the yield is greatly increased, and in the Mond producer, where 2 to 2½ tons of steam are introduced per ton of coal carbonised, the production of ammonium sulphate reaches 90 lb., or some 60 per cent of the theoretical yield. The reasons for these wide divergences will be discussed later.

PRODUCTION OF AMMONIA IN THE THERMAL DECOMPOSITION OF COAL

The amounts of ammonia which are evolved when a coal is carbonised vary with the temperature attained. Anderson and Roberts[1] in 1898 showed that when coal is heated out of contact with air the temperature at which the evolution of ammonia begins depends upon the "age" of the coal in question, and varies from 333° C. for "young" coals to as much as 480° C. for anthracites. In 1908 this result was confirmed by Christie,[2] who showed that, while the evolution of ammonia begins at about 350° C. in the case of gas coals, it is delayed until about 450° C. for anthracites. He stated that the main evolution is found between 500° C. and 700° C., and that beyond this upper temperature little further evolution occurs. Later, Simmersbach[3] gave figures showing the relative amounts of nitrogen occurring in coke, gas, ammonia, tar and cyanogen at different carbonisation temperatures. His figures, which are given in Table XLII, show that ammonia production is a maximum at 900° C., but that even at 1200° C. more than 25 per cent of the original nitrogen is retained in the coke.

TABLE XLII

AMOUNTS OF NITROGEN IN THE PRODUCTS OF COAL CARBONISATION
(Simmersbach)

Temperature of Carbonisation ° C.	Percentage of Nitrogen in				
	Ammonia.	Gas.	Tar.	Cyanogen.	Coke.
600	7·81	18·15	1·12	0·25	71·69
700	18·13	12·13	3·65	0·66	65·43
800	21·28	10·73	3·47	0·87	63·65
900	24·12	12·14	4·15	1·19	58·40
1000	23·15	21·53	4·11	1·23	49·98
1100	23·69	30·51	3·70	1·31	41·39
1200	22·84	45·10	4·21	1·42	26·43

[1] Anderson and Roberts. "The Recovery of Nitrogen in Coal Distillation." *Journ. Soc. Chem. Ind.*, **18**, 1098. [2] Christie. *Inaug. Diss., Aachen*, 1908. [3] Simmersbach. "Untersuchungen ober die Bildung von Ammoniack und Zyanwasserstoff bei der Steinkohlen Distillation." *Stahl und Eisen*, 1914, **34**, 1153, 1209.

Several workers have since given attention to this important subject, and attention may be drawn in particular to the results of Burgess and Wheeler,[1] which are given in Table XLIII. These investigators distilled 2 grammes of coal at temperatures ranging from 350° C. to 900° C. in steps of 100 degrees, and the rapid removal of the products evolved in each stage was provided for. With a Silkstone coal the evolution of ammonia continued up to 900° C. In a repetition of the experiment, this time in a vacuum, the evolution of ammonia began below 350° C. and continued to 600° C., above which temperature no further ammonia was expelled.

The amounts of ammonia evolved in these experiments again represent only a small fraction of the total nitrogen of the coal heated. It must, however, be borne in mind that ammonia is a gas which, under the influence of heat, readily dissociates into its constituent elements. The actual ammonia yields do not therefore necessarily express the amounts formed, but only that which has survived, partial dissociation undoubtedly adversely affecting the yield.

TABLE XLIII

AMMONIA GAS EVOLVED AT DIFFERENT TEMPERATURES
(Burgess and Wheeler)

Temperature ° C.	450	500	600	700	800	900
Total Gas evolved in ccs. per gramme of coal	12·0	29·9	99·0	124·0	218·0	268·0
Percentage of NH_3	4·7	1·35	1·40	1·60	1·00	1·00

DISSOCIATION OF AMMONIA

Ramsay and Young[2] have shown that the decomposition of ammonia gas by heat starts at 500° C., and under favourable conditions, which depend largely upon the area and nature of the surfaces to which the gas is exposed, is practically complete at 800° C. This temperature is, however, that at which in gas-making practice the highest yield of ammonia is produced, as is well illustrated in Table XLIV given by Lewes[3] and as would be anticipated from the work of Christie, Simmersbach and Burgess and Wheeler.

Later work has shown that the explanation of this apparent contradiction is to be found in the retarding action of certain of the constituents of the gaseous products of carbonisation upon the normal rate of dissociation of the ammonia gas.

[1] Burgess and Wheeler. "The Volatile Constituents of Coal." *Trans. Chem. Soc.*, 1910, 97, 1917.
[2] Ramsay and Young. "The Decomposition of Ammonia by Heat." *Trans. Chem. Soc.*, 1884, 88.
[3] Lewes, Vivian. *Carbonisation of Coal* (Benn Bros.)

Recently, Mott and Hodsman[1] have made a study of the decomposition at different temperatures of mixtures of 1 per cent to 2 per cent of ammonia with various other gases, for example coal gas, hydrogen and nitrogen, the experiments being carried out in the presence of a firebrick " chattered " by the attack of salt vapours in a coke oven.

TABLE XLIV

AMMONIUM SULPHATE AT DIFFERENT TEMPERATURES (Lewes)

Temperature. °C.	Ammonium Sulphate. lb. per ton of Coal.
400–500	12
500–600	15
600–700	17
700–800	20
800–900	26
900–1000	24
1000–1100	20

In nitrogen the decomposition of ammonia was found to be perceptible at 500° C., and was complete at 600° C. In hydrogen, however, the dissociation was checked, the same speed of decomposition being reached at temperatures about 150° higher than in nitrogen. For example, at 660° C., when the ammonia was almost completely decomposed in the atmosphere of nitrogen, decomposition in hydrogen was only just measurable.

In a mixture of 1–2 per cent of ammonia with coal gas, 3 per cent of water vapour was found to reduce the dissociation at 700° C. from 17 per cent to 2 per cent, and at 800° C. from about 90 per cent to about 30 per cent. At 850° C. dissociation was completely arrested when about 25 per cent of water vapour was present in the mixture. It was therefore concluded that water vapour exerts a strong preservative action on the ammonia diluted with coal gas.

It has been suggested that as the dissociation of ammonia is closely related to the prevailing temperature, the presence of oxygen may increase ammonia losses by causing local rise of temperature. In Mott and Hodsman's experiments, however, small proportions of oxygen were found to retard dissociation. The explanation of this fact was ascribed to the probable preferential union of the oxygen with hydrogen to form water vapour, the latter having been proved to exercise a strong inhibitory effect upon decomposition of ammonia.

---

[1] Mott and Hodsman. "Factors influencing the yield of Ammonia in the Carbonisation of Coal." *Gas Journ.*, Aug. 15 and 22, Sept. 12, 1923.

AMMONIA PRODUCTION IN AN ATMOSPHERE OF HYDROGEN

Tervet[1] in 1883 passed a current of hydrogen over coke heated to redness in an iron tube, and found that large volumes of ammonia were emitted, the aggregate ammonia yield from the coal being increased by about 100 per cent. From a hard, high temperature coke, however, heating in hydrogen did not produce ammonia. Tervet's results were confirmed and extended by Beilby[2] in 1884.

Further work was described in 1908 by Christie,[3] who found that a current of hydrogen had practically no effect upon the distillation of coal at 480° C., and that further, when a gramme of Shamrock coke was heated at 670° C. in a stream of gas containing 55 per cent of hydrogen, there was no diminution in the nitrogen content of the coke brought about. He concluded, therefore, that the action of the hydrogen was not to cause an increased emission of nitrogen from the coke, but only to diminish the dissociation of the ammonia formed.

Cobb[4] has determined the action of hydrogen upon soft, medium and hard cokes (made respectively at 500° C., 800° C. and 1100° C.) heated successively at 600° C., 800° C. and 1000° C.

During the coking process to 500° C., 21·6 per cent of the nitrogen of the coal had been expelled in various forms, 78·4 per cent remaining in the soft coke. Further heating in hydrogen reduced the nitrogen content in the coke to 28·2 per cent of its original value (22·1 per cent of that in the original coal) ; but of the 71·8 per cent loss which this represents, only 34·2 per cent (or 26·8 per cent calculated on the nitrogen content of the coal) appeared as ammonia, the remaining 37·6 per cent (29·5 per cent calculated on the coal) having been lost chiefly as free nitrogen. The effect of the heating was practically complete at the 800° C. stage, there being little further loss of nitrogen or recovery of ammonia in the stage of heating at 1000° C.

Similar treatment of the medium coke, which retained 49·7 per cent of the nitrogen of the original coal, resulted in a loss of 41·0 per cent of its nitrogen (20·6 per cent calculated on the nitrogen content of the original coal), of which, however, only 3·9 per cent appeared as ammonia. Further experiments showed that the evolution of ammonia was not yet completed, and that prolonged heating would have resulted in more coming off.

With the hard coke, produced at 1100° C., there was no diminution of nitrogen content on heating in the hydrogen atmosphere, and no ammonia was evolved.

[1] Tervet. " On the Production of Ammonia from Coke, resulting from the Destructive Distillation of Coal." Journ. Soc. Chem. Ind., 1883, 3, 445.

[2] Beilby, Sir G. " On the production of Ammonia from the Nitrogen of Minerals." Journ. Soc. Chem. Ind., 1884, 3, 216.

[3] Christie. Inaug. Diss., Aachen, 1908.

[4] Monkhouse, I., and Cobb, J. "The Liberation of Nitrogen from Coal and Coke as Ammonia." Report of Gas Research Fellowship, 1921 ; Trans. Inst. Gas Eng., 1920, 1921, p. 543.

Briefly, then, the action of the hydrogen on the soft coke effected a considerable attack on the nitrogen present in the coke, and there were formed increased quantities of ammonia, recoverable because formed under suitable conditions. With the medium coke the effect of the hydrogen was less marked and the amount of ammonia recoverable was smaller, whilst the hydrogen did not attack the hard coke at all. The rate of recovery of ammonia was always more rapid at 800° C. than at 600° C.

In an extension of this work 94·3 lb. of sulphate of ammonia per ton of coal was obtained by heating a soft coke slowly in hydrogen in stages to 1000° C. Between 800° C. and 900° C. the coke lost 11·7 per cent of its original nitrogen, of which 6·5 per cent appeared as ammonia ; while from 900° C. to 1000° C. only 3·1 per cent of the 9·4 per cent of nitrogen lost from the coke was obtained as ammonia. Even above 1000° C. some ammonia was formed and escaped decomposition.

It was therefore concluded that the presence of hydrogen in the preliminary carbonising period 500–800° C. " in addition to any preservative action in-retarding the dissociation of ammonia once formed, has a specific action in that it is capable of liberating a portion of the ammonia which would otherwise be a stable constituent of the coke."

At 1000° C. the nitrogen in the coke had been reduced to 11·2 per cent of its original value, and was still decreasing, but the rate of emission had become very slow, and was chiefly in the form of free nitrogen.

AMMONIA PRODUCTION IN AN ATMOSPHERE OF STEAM

In his presidential address on ammonia supplies to the Society of Chemical Industry in 1889 Mond remarked that it had been the custom in his laboratory to estimate nitrogen in coke by burning the coke in a current of steam, which converted the whole of the nitrogen present into ammonia. The effect of steaming in increasing the ammonia yield in carbonisation processes is well known.

Following upon their earlier work, Monkhouse and Cobb[1] set themselves to determine what amounts of additional ammonia it is possible to obtain from a coke after the first period of heating to 500° C. is passed, by further heating (1) in an inert gas such as nitrogen, and (2) in such a " potentially reactive " gas as hydrogen or steam, which might be capable of attacking nitrogen compounds with formation of ammonia.

By heating to 800° C. in the nitrogen atmosphere, ammonia to the extent of about 10 per cent of the nitrogen of the coke was evolved, but the total loss of

[1] Monkhouse and Cobb: "The Liberation of Nitrogen and Sulphur from Coal and Coke." Report of Gas Research Fellowship, 1922 ; *Trans. Inst. Gas Eng.*, 1921, 1922, p. 137.

nitrogen from the coke was about three times this amount, the rest being accounted for as free nitrogen. A stream of hydrogen was then substituted for the nitrogen and further ammonia immediately resulted, ultimately equivalent to a further 31 per cent of the nitrogen of the coke. The remaining coke was then subjected to steam, and additional ammonia to the extent of 39 per cent was recovered. Determinations of the nitrogen content of the residual coke after each stage showed that the ammonia production in the last two stages agreed with the nitrogen losses from the coke. It was found that, by submitting the coke to a steam current only, the same final yield of ammonia was obtained. When carbonisation was carried out in steam throughout it was possible, within the errors of experiment, to obtain the whole of the nitrogen of a coal as ammonia, which was liberated much more rapidly than in hydrogen.

When a hard, high temperature coke was heated in steam, the nitrogenous constituents of the coke were attacked, but the liberation of ammonia was far slower than in the case of soft coke ; there was, however, a steady evolution of this gas. Free nitrogen was also shown in the balance sheet, suggesting decomposition of some of the ammonia formed.

NATURE OF THE NITROGENOUS CONSTITUENTS OF COAL

It may be inferred from the results obtained by Simmersbach, Cobb and Monkhouse and others, that the nitrogen in coal is probably present in more than one form. One of the nitrogenous constituents apparently gives ammonia on simple distillation, while the other (or others) is more stable, but is attacked by hydrogen or steam with formation of ammonia.

Christie suggested that coal contains " imino " nitrogenous compounds which decompose below 900° C. yielding 'ammonia; and very stable " nitride " forms, which remain in the coke after ordinary carbonisation, but which are gradually expelled as free nitrogen when the temperature is raised from 900° C. to 1900° C.

Cobb suggests that there is indication of " amino " groups, which give ammonia by simple distillation, and more stable compounds which are possibly complex ring formations with nitrogen existing in the ring.

PRODUCTION OF AMMONIA IN GAS WORKS PRACTICE

In normal gas-making the primary formation of ammonia occurs by the decomposition by heat of nitrogenous organic substances in the coal, followed by its secondary formation at higher temperatures produced by the action of the hydrogen in the coal gas upon the soft coke at a red heat. This ammonia, as the temperature rises, would undergo more and more dissociation, and would consequently be evolved in ever-decreasing quantities, until at about 800° C. the

production would be nil, were it not for the checking action of the hydrogen present upon the dissociation process, which, as has been shown, exercises a marked influence upon the ammonia yield.

The explanation of the effect of steaming in vertical retorts on the ammonia production becomes clear from the previous considerations. The advantages of steaming for high temperature carbonisation are unquestionable. It is now clearly established that increased therms in the form of gas and increased quantities of tar and ammoniacal liquor are to be obtained by this method, although the quantity of coke is reduced. In regard to the production of ammonium sulphate, full-scale experimental work at H.M. Fuel Research Station[1] produced the results given in Table XLV.

TABLE XLV

EFFECT OF STEAMING ON PRODUCTION OF AMMONIUM SULPHATE

Coal.	Percentage of Steam.	Ammonium Sulphate. (lb. per ton of coal.)
Consett	Nil.	14·65
	5·24	17·30
	20·95	20·00
Mitchell Main Gas Nuts	Nil.	16·27
	5·06	18·75
	12·44	21·8
	20·53	21·2
	30·03	21·95
Main and Ell, Lanarkshire	Nil.	32·6
	22·95	43·9

Salmang[2] has stated that the ratio of carbon to nitrogen in coke remains constant whilst gasification in steam is taking place. If this observation is substantiated, only limited steaming could be used for increasing ammonia production in such processes as are designed to yield coke as a principal product. Cobb has suggested that at each temperature stage in steam there is probably a double reaction taking place : (1) a decomposition of ammonia yielding compounds in the coke such as accompanies a rise of temperature even in an inert atmosphere of nitrogen ; and (2) a reaction also liberating ammonia, but accompanied by and probably conditioned by the gasification of the carbon of the coke.

[1] Fuel Research Board. Report for Years 1920, 1921 : First Section : " Steaming in Vertical Gas Retorts." (H.M. Stationery Office.)
[2] Salmang. " Dissertation der Technischen Hoehschole Aachen," 1914. See, Markgraf "Ammonia Formation in the Gasification of Coke and Coal by Steam and Air," Journ. Soc. Chem. Ind., 1916, 35, 366 ; Stahl und Eisen, 1915, 35, 905,

YIELD OF AMMONIA IN LOW TEMPERATURE CARBONISATION

In view of the widely divergent, and in some cases extravagant, claims of some inventors for the yield of ammonium sulphate by their respective processes, it has been considered desirable to discuss at some length the latest researches in the nitrogenous products from coal. It is evident from what has been said, that in carbonisation processes so effected that no part of the coal is heated to temperatures higher than 600° C., the yield of ammonium sulphate from bituminous coal will be of the order of 15 lb. per ton, though even in this case, if the products are subjected unnecessarily to the full temperature of the retorts for any considerable time, a proportion of the ammonia will be decomposed in accordance with the results of the investigations of Ramsay and Young and Mott and Hodsman. At such comparatively low temperatures, however, such losses are not usually very considerable, the greater part of the nitrogen of the coal still being retained in the coke.

In processes where carbonisation is carried out in stages, it is possible, without altering the nature of the products first driven off, to secure increased aggregate yields of ammonia by the subsequent attack of the hydrogen in the coal gas liberated upon the nitrogen in the hot coke, and its further action in retarding dissociation. For example, in the " Pure Coal Briquette " and the " Sensible Heat " (Nielsen) systems, where the temperature is graded in such a way that the coke first formed passes through successively increasing zones of temperature, the amount of nitrogen liberated as ammonia is continually augmented as the temperature rises. The fact that the gases escape via the cooler end of the retorts tend to preserve this additional quantity of ammonia.

A readily available way of determining the possible yield of ammonia from coal is provided by the assay method already mentioned. The yields of ammonia from various coals as given by this method have been correlated with those obtained from the same coals in horizontal retorts at H.M. Fuel Research Station, and have been found to be substantially the same.[1]

In some of the proposed manufacturing processes the yields quoted are extremely high when the above considerations are taken into account. Some of these claims may be justifiable in those cases where the conditions favour the production of ammonia by the secondary reaction, but in the majority of cases it is extremely unlikely that yields exceeding 12 lb. of sulphate of ammonia per ton of coal can be obtained. In both the assay apparatus and the horizontal steel retorts of H.M. Fuel Research Station the yields varied from 5 to 10 lb. per ton according to the type of coal carbonised.

[1] Fuel Research Board. Report for Years 1920, 1921 ; second section : " Low Temperature Carbonisation." (H.M. Stationery Office.)

N

In a normal bituminous coal containing 5 per cent moisture, the yield of liquor per ton of coal would be approximately 10 gallons from the coal itself, and 11 gallons from the moisture, or a total of 21 gallons.

In order to pay for recovery, this liquor would require to be of at least 8 oz. strength or about 1·7 per cent ammonia. The liquor as collected would therefore have to show a recovery of 14 lb. per ton to reach this strength. Otherwise it would be necessary to circulate the collected primary liquid in an ammonia scrubber until it had reached the desired strength of 1·7 per cent ammonia. It is practically certain that only a small proportion of the total liquor formed could be concentrated in this way, and in view of the small yield available for sale and the low prices ruling at the present time, it will be seen therefore that ammonia recovery is hardly worth taking into account in the balance sheet of a straight low temperature process.

Apparently no special examination of the distribution of other nitrogenous bodies or of sulphur compounds in low temperature liquor has yet been carried out ; but these will probably bear some similarity to those found in normal gas. liquor, although their distribution will certainly be different.

CHAPTER XVI

PROCESSES—EARLY WORK

INTRODUCTORY

ALTHOUGH it had been known for many years that the distillation of coal yielded an inflammable gas, it was not until the close of the 18th century that coal gas was utilised for the production of light and heat. This was due to the pioneer work of William Murdoch, who may rightly be considered the founder of the modern gas industry. Murdoch's experimental work, begun in Cornwall in 1792, and continued in the Soho works of Messrs. Boulton and Watt, of Birmingham, in 1799, was so far successful that the gas was used in connexion with an outdoor illumination to celebrate the Peace of Amiens in 1802. In 1810, the first Act of Parliament was obtained for the formation of a Statutory Company to supply coal gas to the public ; and since that date the industry has developed by leaps and bounds until its ramifications are now world-wide. It is unnecessary in this volume to trace the various stages of development of the gas industry. Even now finality has by no means been reached, and owing to the freedom in the selection of the most suitable methods of working afforded to gas engineers by the Gas Regulation Act of 1920, it is at present somewhat uncertain in what direction the high temperature carbonisation process will tend to stabilise itself in the future.

Compared with high temperature carbonisation, the process of low temperature carbonisation has had a very short history. Although the first gas-making processes were carried out under low temperature conditions, the trend of development has always been in the direction of obtaining increased gas yields by the use of increasing temperatures, and over a hundred years elapsed from the time of Murdoch's early experiments before the possibilities presented by the carbonisation of coal at low temperatures were recognised. It is true that sporadic attempts were made to initiate work of this description, but in the main, inventors who turned their attention to the carbonisation of coal have usually worked more with the idea of increasing the yield of gas and decreasing the yield of tar than with that of obtaining solid smokeless fuel and a plentiful supply of liquid fuel. The liquid fuel would indeed have been a drug upon the market in those days, since it is within comparatively recent times that oil-burning appliances for steam-raising and industrial furnaces and the internal combustion engine have been developed. The proposal of the process of low temperature carbonisation has been made firstly, for obtaining a smokeless fuel, and secondly, for the increase of supplies of home-produced liquid fuel to meet the increasing demands of industry during the twentieth century.

COALITE PROCESS

For the process of low temperature carbonisation, as it is understood to-day, there is little doubt that credit must be given to Mr. T. Parker, of " Coalite "

fame. Parker's first patent was granted in 1890, but his principal patents for the process were taken out in 1906. In the years 1907 and 1908 numerous other patents were filed in his name. Experiments were first conducted in ⌂-shaped horizontal retorts ; but, after experimental work on various other types, Parker devoted his energies to the development of the process in cast iron vertical retorts, about ten feet long, tapering in diameter from about four inches at the top to about six inches at the bottom. Twelve tubes were cast together in a nest, and heated externally so that the material reached a temperature of about 450–500° C. The low temperature coke, known as coalite, was found to be admirably suitable for a domestic fuel, and great commercial success was expected from the plant.

These sanguine expectations were not, however, realised. A further study of the published records reveals the great difficulties with which Parker had to contend in putting his process upon a commercial basis. For example, he took out patents on the improved arrangements for withdrawing gas in the event of any retort becoming choked at its upper part and for discharging the coke after carbonisation—troubles which have not been entirely overcome by the inventors of other methods of low temperature carbonisation. The death of Parker undoubtedly retarded the development of this process. The later retorts were made of firebrick, and had a rectangular cross-section, very wide in relation to the breadth. Four zones of heating were introduced, the lowest at 450° C., the next at 500° C., next at 550° C., and the highest at 900–1000° C. The function of this high temperature at the top of the retort was to crack the tars, to facilitate which a grid of suitable material and shape was suspended in the free space. This retort, like its predecessors, was not very satisfactory, and further experimental work had to be undertaken. Low Temperature Carbonisation, Limited, the successors to the original company, and the holders of the Parker patents, have evolved a retort which is now being tested on a commercial scale at Barugh, Barnsley, and which is described on page 206.

OTHER PATENTS

In dealing with the work of the earlier pioneers, it is not possible to describe the many inventions which have been patented, or the many experimental plants which have been built. It is proposed, therefore, in the remainder of this chapter, to describe the pre-war and war-time developments and contributions to the technique of the subject, leaving for a further chapter the post-war developments which have taken place.

RICHARDS AND PRINGLE RETORT

In 1909 a further method of carbonising material at low temperatures was invented by Richards and Pringle. This process was continuous in action, the

material being run into V-shaped buckets on an endless conveyor, which passed through a horizontal retort heated externally. The advantage of this process was that the material was undisturbed during coking, and a very satisfactory coherent fuel could be obtained. As the material was also in small charges, the time of carbonisation was short, and a large throughput might be expected. Difficulty was at first experienced in discharging the coked material from the buckets, though this was eventually remedied to a large extent. The apparatus is, however, bulky for the throughput obtained, and the possibility of the loss of gas through leakage is therefore increased. Other difficulties, due for example to the pin joints of the conveyor being subjected to high temperatures, have been encountered, and much credit is due to the inventors for their praiseworthy attempts to make this scheme ultimately successful.

TOZER RETORT

In 1909, also, Mr. C. W. Tozer patented a further process intermittent in action. The distinctive features were the use of a high vacuum system, previously patented by W. S. Simpson, and a special design of circular vertical retort. The retort is of cast iron, and has its cross-sectional area divided by concentric rings connected by webs, which extend the whole length of the retort, so that the material is charged into annular spaces about four inches wide, as shown in Fig. 31. Due to the conductivity of the webs, the diameter of the retort, and hence its capacity, can be increased considerably over that of a plain tube without danger of reducing the uniformity of coking of the charge, while the shape and the material of the retort enable it to withstand the pressure due to the expansion of the charge, thus ensuring the production of a dense, homogeneous smokeless fuel. It is stated that experience extending over a number of years has shown that cast iron retorts for low temperature carbonisation are practically indestructible. The time for retorting of the charge is about 4 to 4½ hours. The central core is sealed with a stopper during charging, and being uncharged facilitates the withdrawal of the gases and oil vapours by forming a gas passage connecting the lower to the upper part of the retort. The gases evolved from the charge partly pass upwards through the carbonising mass and partly downwards. Those gases which pass downwards make their exit by
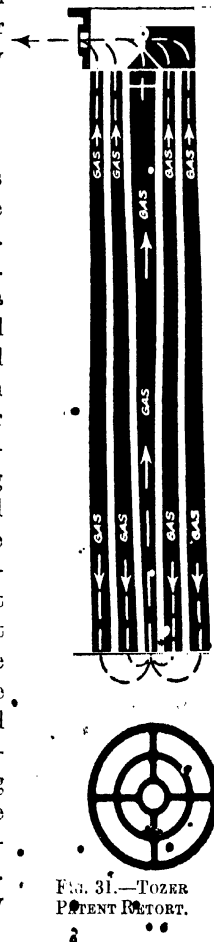
Fig. 31.—Tozer
Patent Retort.

way of the central tube to the eduction pipe which is situated at the top of the retort. The double exit for the gases, together with the partial vacuum which is maintained in the system, enable the gases to be removed rapidly from the retort. The retort was originally kept under a vacuum of 20-27 inches of mercury, but this high vacuum is not now specified, being reduced to 70-150 mm. of mercury. The charging is effected through a top door ; and when the carbonisation period is completed, the coke is discharged through the bottom door of the retort. The charging and discharging doors are made tight by a special arrangement, and the lower door is balanced by counterweights. The doors are easily locked. The retorts are heated by a specially designed recuperative setting. Each retort is surrounded by a ring of brickwork to prevent flame impinging direct on the cast iron casing, and the space between the retort and brickwork acts as a heating flue. In each of the exit gas flues is a damper which controls the temperature of the retort. After passing through the damper, the exit flue gases make a number of passes in a recuperative flue, the secondary air for the combustion of the heating gases being passed through boxes at right angles across the flue. It is claimed that there is rarely trouble in getting the coke out of the retort. Illustrations of the experimental plant at Battersea are shown in the *Engineer* of 23rd September, 1921, and a description of the retort which was being constructed at the Fraser and Chalmers Engineering Works is also given.

MARYHILL RETORT

The problem was also investigated experimentally by Mr. G. T. (now Sir George) Beilby, at the Maryhill works of the Cassel Cyanide Company, Glasgow, and the position at that time is given in a contribution to the discussion on " Fuel Economy," held in Section B of the British Association at Birmingham, 1912, which is reproduced in full in the Report of the Fuel Research Board.[1] The retort, patented in 1910 by Mr. H. N. Beilby, consisted of an externally heated vertical shell, containing a series of plates which were inclined alternately in opposite directions. The material was passed through the retort in layers about two inches thick by jerking or vibrating these shelves. The maximum temperature to which the material was subjected in this apparatus was 400-450° C., and the time of carbonisation was reduced from the four to six hours necessary in the Parker retort to about one and a half hours. The reduction was due, not merely to the treatment of the material in thin layers and the free escape of the volatile gases, but also to the frequent turning over of the coal as it passed over the shelves, and the consequent exposure of fresh surfaces to the action of the heat.

As the resulting coke was in small pieces, briquetting was necessary in order

[1] Fuel Research Board. Report for Years 1918, 1919 (H.M. Stationary Office.)

to produce a fuel which was suitable for domestic purposes. The process also had the disability of being able to treat non-caking coals only. Although improvements were effected so that a unit was eventually obtained in which a throughput of fifteen tons per day was easily maintained, it was decided to discontinue the experiments. It was considered that a retort of this description was too limited in its range of application to provide a solution of the wider national problem of the satisfactory treatment of fusible, or partly fusible coals, at low temperatures. The experience gained in the work has since been advantageously employed in the design of low temperature carbonisation plant at H.M. Fuel Research Station, which will be later described.

DEL MONTE AND CHISWICK PROCESSES

Of the continuous processes, one of the best known is the Del Monte, embodied in what has come to be called the Chiswick retort. The development of this process can be followed very clearly by an examination of the patents which have been obtained by those in charge. The first patent was obtained by Del Monte in 1911 and the process for a considerable time went by that name. This patent relates to the extraction of the volatile matters in carbonaceous materials by the diffusion through the heated material of a gaseous medium which is preheated to the same, or to nearly the same, temperature as the charge within the retort. This idea was embodied in a small vertical intermittent type of retort, the gaseous medium employed being coal gas. Apart from other non-commercial aspects of a plant of this description, the results were unsatisfactory, because of the great difficulty of ensuring a uniform diffusion of the hot gas through the charge. A radical change in the method of retorting was therefore made, and in 1914 a patent was obtained in the name of the Oil and Carbon Products, Ltd. According to this patent, the retort was given a small inclination to the horizontal, and the material was moved through and up the retort by means of a screw conveyor of almost the same size as the internal diameter of the retort. The pitch of the screw and the diameter of the retort were kept small, so that the material did not move through the retort in bulk, but was heated in relatively thin layers. The retort was heated externally by gas, or according to a separate patent filed the same year it could be additionally heated by means of gas jets placed within the conveyor shaft, which was made hollow for the purpose. The process was therefore continuous in action, and represented a great advance upon the previous patent. This patent was obtained not so much upon the method of retorting—for of course the principle of the screw conveyor is old—but on the method of condensing the products of distillation. The raw material was fed in at the lower and colder end of the retort, and the retort was progressively heated towards the upper end. The oil and gas outlet

being at the lower end of the retort, the hot gases and vapours were condensed by contact with the slowly progressing colder material passing up the retort and the condensates were thus easily collected.

It was found, however, in practice that the oils were so heavily laden with free carbon and ash that they were commercially of little use. A possible remedy is reflected in the next patent, dated 1915, in which the retort is horizontal, gaseous products of distillation being collected at different points in the length of the retort by separate eduction pipes. These eduction pipes lead the gaseous products into a collector, which likewise largely acts as a condenser. The gas outlet being at the cool end of the collector, the patent is obtained on the claim that the hottest vapours have to pass through successively cooler zones of vapour on their way to the outlet.

Up to August, 1918, no further patents had been obtained and the improvements effected had been of a minor character. In 1917, the Petroleum Research Department of the Government ran a large number of tests upon the plant. These tests were made, however, more for the purpose of determining the oil values of various materials than of proving the commercial possibilities of the process. The tests were so far successful that, under an agreement with the Mineral Oil Production Department of the Ministry of Munitions, a battery of thirty retorts was under construction and nearly completed when the armistice was signed. In consequence of the changed conditions the work was temporarily stopped, and the plant was afterwards dismantled. Each retort was 14 in. diameter, and about 20 feet long, and would have had a throughput of about $1\frac{1}{2}$ tons per day. The name of Del Monte has now been largely dropped in connexion with this plant, and the Chiswick retort, as it is now called, occupies a prominent place in the history of the development of the low temperature system of carbonisation.

It should be noted that the Chiswick retort compares with the proposed Burney retort, patented in 1918. The Burney retort is about 6 feet in diameter, and about 30 feet long. As in the Chiswick retort, the material is moved through the retort by means of a screw, but in the Burney retort the screw is hollow, and carries the hot flue gases. In this way not only can the diameter, and consequently the throughput, be increased, but the carbonising heat is economically and efficiently utilised. A description of the Burney retort and process is given elsewhere[1] and need not be recapitulated, since a full-scale plant has not yet been built and tried.

MACLAURIN RETORT

In 1909, on the motion of Councillor W. B. Smith, a sub-committee of the Glasgow Corporation Gas Committee was appointed to " Inquire into and report

[1] McKey, R. F. *The Burney Retort for the Treatment of Carbonaceous Material.* (Waterlow.)

on any process of gas-making that will produce a form of coal residue that may
be available for use in ordinary domestic grates." In 1914 all the known processes
for the production of smokeless fuel were examined, and it was decided that the
process patented by R. Maclaurin[1] in 1913 seemed the most promising for this
purpose. As this process also promised to yield large volumes of gas at a low price,
Bailie Wm. B. Smith moved, in March, 1914, for the appointment of a sub-
committee of the Electricity Department to work along with the sub-committee
of the Gas Department on smokeless fuel, with the object of deciding whether
gas could be utilised economically for the firing of boilers. Negotiations were
entered into and proved successful, and in 1915 a plant to carbonise 20 tons
of coal per day was erected at Port Dundas, near Glasgow.

The Maclaurin process differs radically from any of those preceding, inasmuch
as the fuel is carbonised in an internally heated retort, as opposed to the externally
heated types previously described. The retort resembles an elongated producer
or blast furnace, but is on a smaller scale.

According to the original patent, the inventor proposed to pass heated gas
through a column of bituminous fuel in a high producer, the temperature of which
decreases upwards, and to collect the condensed oils within the upper part of the
producer, so as to prevent them from running back to the lower and hotter part
of the column.

The first producer was put down in 1914 at a small colliery in Ayrshire, with
the object of treating cannel coal and making gas for boiler firing. The coal,
having a high ash content, was burned to ash. This work proved the soundness
of the principle of trapping the oil in the upper regions, but had to be discontinued
owing to the colliery getting into difficulties. The Glasgow Corporation, however,
had by this time decided to proceed with the erection of a plant primarily for the
manufacture of smokeless fuel. With the object of getting as clean a fuel as possible,
it was decided to erect a double producer. This producer was described by the
inventor in a paper read before the Society of Chemical Industry in 1917.

At that time the plant, which was situated at Port Dundas, near Glasgow,
consisted of two retorts, A and B (Fig. 32). The fuel in the small producer B was
completely burnt to provide power gas, which passed through the mass of coal in
the large producer A, and carbonised it to smokeless fuel. Thus the ash from the
fuel burnt to provide the heating medium did not mix with the smokeless fuel
itself. In the latest illustration of the plant worked at Grangemouth it appears that
a single producer is now being used. This producer differs from the original type
in that the coke does not pass through a water seal at discharge. It is described
more fully in the next chapter, which deals with the later developments.

[1] Maclaurin, R. "Low Temperature Carbonising and some Products." *Journ. Soc. Chem. Ind.*
36, 12.

The Maclaurin plant can deal with a variety of coals. As with other producers, however, it does not work satisfactorily with dross. It is claimed that, owing to the gradual heating, little binding together of the coke takes place except with fairly strongly coking coals. The blast can be readily controlled so that the charge may be entirely gasified, and ash only withdrawn from the bottom, or the coke produced may range from a hard grey fuel suitable for metallurgical purposes to an easily ignited smokeless fuel adapted for domestic use. It is stated that, during carbonisation, any stony matter in the coal tends to drop out, while mineral
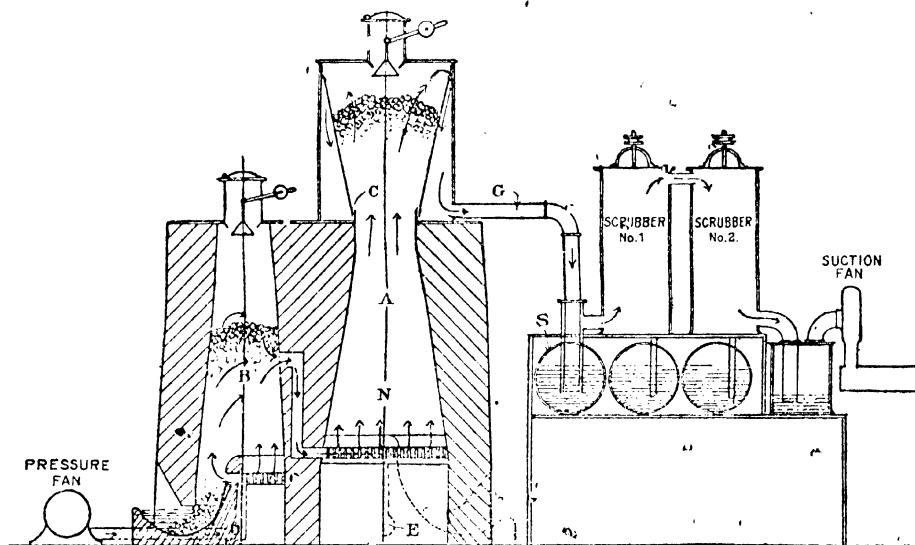
FIG. 32.—MACLAURIN PRODUCER (1917).

matter scales off and is concentrated in the breeze. It is claimed, therefore, that it is possible to obtain a clean coke containing a percentage of ash which is little higher than that of the original coal since the bulk of the ash is separable as stones or segregates in the breeze.

When the experimental work was stopped at Port Dundas, owing to difficulties created by the war, the principal conclusions reached by Mr. Maclaurin as a result of that work were :—

(1) Coal could be turned into smokeless fuel by the passage through it of from 13,000 to 20,000 cubic feet of hot power gas. This is much less than was generally believed to be necessary.

(2) An easily kindled smokeless fuel could be obtained, containing not more than 4 per cent of volatile matter.

(3) In a true low temperature carbonisation the yield of rich gas was very small, being probably not more than half what was generally believed.

(4) The oils obtained were highly complex bodies, difficult to investigate by distillation, as they appeared to suffer decomposition on re-distillation, giving rise to new products.

(5) Ammonia was obtained in fair quantity, but, while the liquors were free from ferro-cyanides, sulpho-cyanides, etc., they always contained certain complex polyhydric phenols, capable of dyeing iron-mordanted wool a deep slate grey.

(6) The most important conclusion of all, however, was that a plant of this type could work with coking coals without sticking up, a difficulty which had been almost universally predicted by all those experienced in coal carbonising.

The plant at Port Dundas was taken down and re-erected at Grangemouth, where further experimental work is now proceeding. The process appears to be one of great promise. The first cost, and the cost of maintenance, are low ; the labour costs are also low, and the heating arrangements efficient. It still remains to be proved, however, that the gas, oil and coke will find ready and profitable outlets when a plant on a commercial scale has been installed in some centre of industry.

ESTABLISHMENT OF H.M. FUEL RESEARCH STATION AT EAST GREENWICH

In addition to the above processes initiated and developed during the pre-war and war-time periods, numerous other patents had also been taken out at the same time, and many other experimental plants built. Interesting though many of these may be, it is not possible within the confines of a single volume to give an extended description of all. During this period, however, it was becoming more and more apparent that the difficulties of low temperature carbonisation were more profound than had been anticipated by the early pioneers ; and that for the development of each and every process some measure of co-operation and mutual assistance was essential between those interested in the solution of the problem. This development was in reality part of a larger movement. For many years there had been a growing consensus of opinion that a special need existed for some degree of State organisation and assistance in every form of scientific research. It was felt that if this country were to advance, or even maintain its industrial position in the markets of the world, there must be such a development of scientific and

industrial research that the national industries could expand naturally and compete successfully with those of other highly organised countries. These views finally took form when, in July, 1915, a Committee of the Privy Council for Scientific and Industrial Research appointed an Advisory Council composed of men eminent in science and industry. One of the primary functions of this Advisory Council was to advise on " proposals for establishing or developing special institutions or departments of existing institutions for the scientific study of problems affecting particular industries and trades.".

It was soon recognised that certain lines of investigation existed which affected so large a number of interests and of which the prosecution was, so vital to the well-being of the population that they could not be effectively followed by the mere affording of monetary or other assistance to any particular industry for co-operative research. For example, the question of research into fuel and its utilisation was too comprehensive to be dealt with by any one industry. Accordingly in 1917, a Fuel Research Board was established, and a Director appointed immediately responsible to the Lord President, in order to organise and carry out research investigation on fuel problems.

OBJECTS OF THE FUEL RESEARCH BOARD

It has already been pointed out in Chap. I that the Fuel Research Board have kept in view two main lines of research : First, a survey and classification of the coal seams in the various mining districts by means of chemical and physical tests in the laboratory, and second, an investigation of the practical problems which must be solved if any large proportion of the raw coal at present burned in its natural state is to be replaced by the various forms of fuel obtainable from coal by carbonisation and gasification processes.

In their second Report to the Lord President, the Board stated that :—

"The second of the proposed lines of inquiry has been led up to by a variety of influences during the past eight or ten years. Among these influences have been the demands for cheaper and more ample supplies of electrical energy, for home supplies of fuel oil for the Navy, of motor spirit for the Transport and Air Services, and last, though by no means least, for smokeless domestic fuel. This last has been brought about through the growth of public and municipal opinion on the subject of smoke prevention in cities and in industrial centres.

" The only development which would satisfy all these needs simultaneously would be the replacement of a large proportion of the raw coal which is at present burned in boilers, furnaces and domestic fires, by manufactured fuels prepared from raw coal by submitting it to distillation.

" The greater part of the coal which is consumed in Great Britain is burned

in its natural state as it comes from the mines. The question of the moment then is : to what extent can and ought the present use of raw coal be replaced by the use of one or other of the various forms of fuel manufactured from coal— coke, briquettes, tar, oil or gas. While there is already in the possession of experts a certain amount of knowledge and experience which might enable them to organise and direct schemes for the replacement of raw coal by manufactured fuel in particular directions and on a fairly large scale, no really comprehensive scheme can be formulated till certain perfectly definite problems in coal distillation have been solved.

" These problems can only be solved by carefully organised experiments on a working scale carried out under the conditions likely to arise in practice."

After pointing out the fundamental differences between gas and coke-oven industries already established, and a possible new industry which would provide for the desiderata already referred to, the Report continues :—

" As regards the carbonisation of coal at low temperatures there is no corresponding body of experience in existence and there are very few properly accredited data available."

It will thus be seen that, although the programme of the Fuel Research Board includes the whole question of the more efficient utilisation of fuel, the subject of low temperature carbonisation occupies a foremost place.

H.M. Fuel Research Station at East Greenwich is designed and equipped for the purpose of conducting operations both on laboratory and full industrial scales. Since its opening a large amount of reliable data has been obtained and made available in connexion with the methods of treatment of coal. Although it cannot be claimed that a complete industrial solution of the problems of low temperature carbonisation has been arrived at, either here or elsewhere, it is believed that knowledge and experience have been gained which have brought a commercial solution appreciably nearer. The Board have also, from time to time, assisted other investigators and firms in their experimental work, both by free discussion of the difficulties and possible solutions and, in some cases, by the provision of financial assistance for the erection of plant possessing some specially promising feature.

CHAPTER XVII

PROCESSES—SOME LATER DEVELOPMENTS

INTRODUCTORY

USEFUL though it be to study the work of the early pioneers of low temperature carbonisation, it yet remains to be seen how their failure and successes are reflected in the processes now being considered and actively advocated. It is desired in this chapter to describe and illustrate these processes and retorts (which, it should be added, are placed in alphabetical order, except for a few which are described in tabular form on p. 235).

THE BARNSLEY PLANT

The plant of Low Temperature Carbonisation, Limited, at Barugh, Barnsley, consists of twenty retorts, built as one unit, and has a throughput of about 36 tons per day. Each retort is made of firebrick, cemented with fireclay, and is of the vertical, intermittent, externally heated type. The total depth of the retort is 9 feet 6 inches, the width at the top 7 feet 3 inches, the width at the bottom 7 feet 6 inches, and the breadth 11 inches. The distance from centre line to centre line of adjacent retorts is 21 inches. It is stated that, in a retort of these dimensions, 12 cwt. of coal can be carbonised at a time, and that the time of carbonisation is less than eight hours. The throughput for each retort is therefore about 36 cwt. per day. Each retort hopper carries one charge, and thus acts as a measuring chamber. Sections of the retort are shown in Figs. 33, 34 and 35.

The distinctive feature within each retort is a pair of collapsible iron plates which are suspended about three and a half inches from each side wall. The charge to be carbonised lies between the plates and the walls. The plates are perforated, and the free central space between them is in communication with the exhauster. The function of the plates is twofold. In the first place, the space between the plates forms a central chamber whereby the removal of the gases from the retort is facilitated. This is beneficial in two ways. It reduces the contact of the vapours with the hot charge to a minimum, and as the pressure within the retort is thereby kept fairly uniform, there is less liability for the retort to choke and cause the rich gas to leak through the walls to burn with the flue gases. The other function of the plates is to prevent the jamming of the charge in the retort during carbonisation, and to facilitate the withdrawal of the coke when the bottom door is opened. As shown in Fig. 36, the plates are suspended from a bridge piece in the retort. The partial rotation of the bridge piece moves the plates towards each other, and the coal space in the retort is thus increased. The expanded and collapsed positions of the plates are shown in the same figure.

During charging and carbonising the plates are on the same level. The bridge piece covers the space between them, and deflects the charge into the two outside

FIG. 33.

FIG. 34.

Conveyor

C. of Ram

Coal Hopper

C.L. of Ram

C.L. of Conveyor

Coal

Top Filling
Door

Platform

Operating Levers

Poppet Door

Cooling
Chamber

Waste Heat
Flue

Coalite Retorts
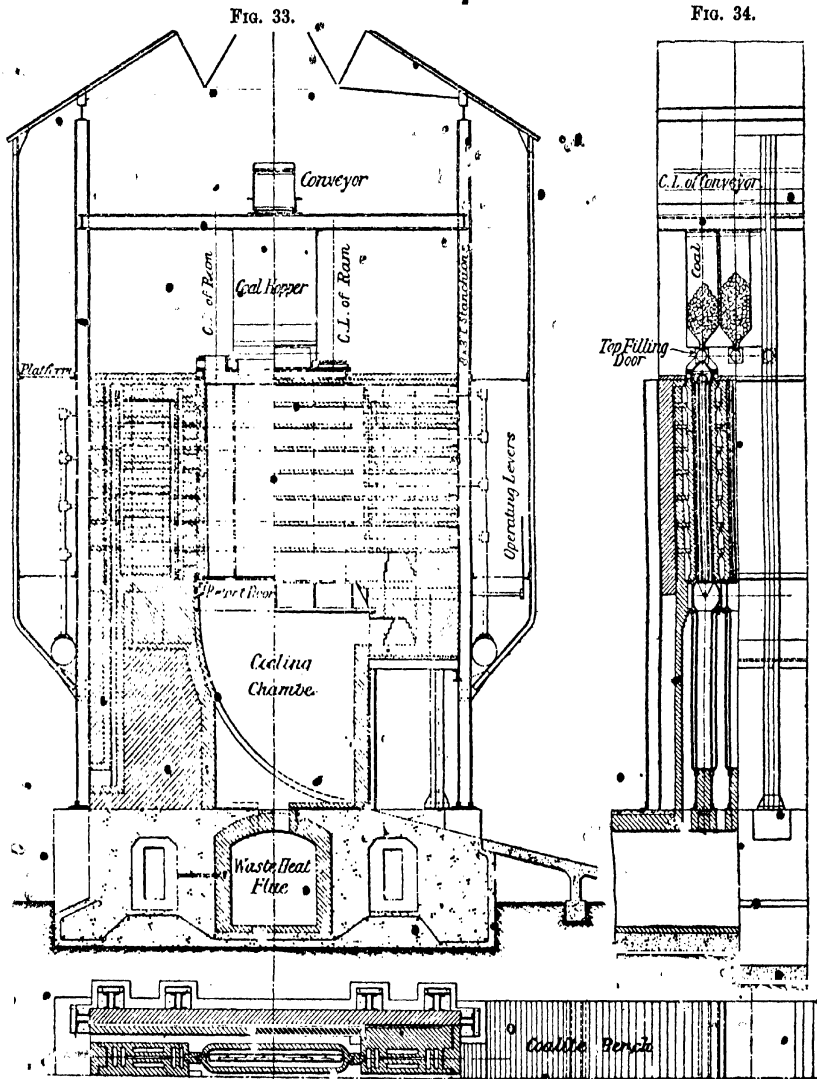
FIG. 35.

FIGS. 33–35.—BARNSLEY RETORT AND SETTING
(Low Temperature Carbonisation, Ltd.).

spaces. The plates are kept apart by flanged abutments, which take the thrust when the coal swells during carbonisation. At the bottom the plates are connected by a link, which determines their maximum separation. When the bridge piece is partially rotated, the plates come together, one plate rising and the other falling by equal amounts, so that the distance between their outside surfaces is reduced from five inches to about three inches. On account of this extra space, the discharge of the fuel after carbonisation is facilitated, and the retort can be readily cleared. These plates are made of manganese cast iron, and they are stated to withstand the somewhat arduous service without suffering undue growth or other trouble.

A further distinctive feature of this plant is the cooling chamber beneath each retort. When the dumping door is opened, the charge drops directly into this chamber. The front of each chamber is closed by an airtight door, through which the fuel, when cooled, is drawn out on a sloping platform extending throughout the length of the retort bench. The cooling chamber is made of sheet steel, the two sides being water-jacketed. The back is curved, to facilitate the clearance of the chamber. Thin vertical plates are fixed centrally in the water jacket, in order to ensure a continuous circulation of the water. It is claimed that practically the whole of the sensible heat of the fuel is thus absorbed, and that the coke can be sufficiently cooled for withdrawal within two hours. The steam generated in the cooling chamber is intended for use in the fractionating of the tar oils.

Steam is admitted through the dumping door, and it is claimed that the action of the steam on the charge undergoing carbonisation results in the increase of the ammonia compounds in the gas given off.

Fig. 36.—BARNSLEY RETORT. Showing collapsible plates.

Top Doors

Sectional View of Plates

End Elevation of Plates

The gas leaves the retort at the top and passes into a hydraulic main through swan necks. The tar extraction plant, gas scrubbing plant, etc., are those of ordinary gasworks practice.

The gas retorts are heated externally by gas obtained, either from the carbonised charge, or from a Mond producer gas plant. A section through the retort flues is

shown in Fig. 33. The burner flues are situated one above another horizontally in the space between the retorts. It will be seen that two large gas mains run horizontally at the front and the back of the retorts and feed the burners through vertical pipes. The burners are plain horizontal pipes. The hot gases of combustion from the top burner pass downwards, regenerated at each successive flue by supplementary burners. The waste gases eventually pass through recuperators and pre-heat the air for the burners.

The charge is a mixture of coking and non-coking coals in the proportion of about 30 : 70, and the mixing occurs while the coal is being washed. The washed slack carries from 10 to 12 per cent moisture. The charge per retort of 12 cwt. produces about 917 lb. of saleable coke, of which not more than 4 per cent is breeze.

THE CARACRISTI PROCESS

The Caracristi process, which has recently been adopted by the Ford Motor Company in the United States, consists essentially of a molten lead bath, over which roughly pulverised coal is passed in layers on a metal plate conveyor. It is stated that a Caracristi retort with a throughput of twenty-five tons per day has been successfully working for two years in America. The chief feature in this process is the short time required for the carbonisation of the material. It is claimed that a layer of coal half an inch thick, passing over a bath of molten lead at a temperature of 650° C., is completely carbonised in five minutes.

Details have been published of a plant now under construction at the Ford Works in Canada, at Walkerville, Ontario. The firebrick retort is about 50 feet long. Gas is the source of heat for maintaining the molten lead bath at the required temperature, and the products of combustion pass through narrow passages under the bath into chequer brickwork regenerator settings overhead. A layer of coal about half an inch thick is run on the continuous metal conveyor which travels on the surface of the lead, and returns under the bath through a tunnel. This funnel also contains a secondary conveyor which is used to cool the coke before it is discharged. The main conveyor consists of cast iron plates which are connected by steel pins and have overlapping joints, so that the coal does not fall on the lead. The speed of the conveyor is such that, as previously stated, the time of carbonisation of the fuel is about five minutes.

It is stated that the normal quantities of the products of carbonisation are obtained by this process, and that the coke contains 12-15 per cent of volatile matter. It is not, however, in a form suitable for use as a domestic fuel when it comes from the retort. It is the intention of the promotors to pulverise the greater proportion of the fuel obtained from the plant under construction, and use it for steam-raising purposes.

o

THE CARBOCOAL PROCESS

The Carbocoal or Smith continuous system of carbonisation, developed in America by the International Coal Products Corporation, should strictly be ranked amongst the high temperature systems but for the fact that the inventor, Charles H. Smith of New York, has attempted to solve the problem in America which corresponds to the problem attacked by low temperature carbonisation in this country.

Coke, which is known as "carbocoal," and oil are the primary products of carbonisation, and gas is a secondary consideration. The process differs from other processes in being in two stages ; the material first being treated at a low temperature, and the resulting semi-carbocoal again treated at a high temperature. A plant working on this system at South Clinchfield, Virginia, has a capacity of 550 tons of carbocoal daily. Another plant has also been constructed at Irvington, New Jersey.

It is possible to carbonise a wide range of materials by this process, including lignite, coking coals and non-coking coals. The material is first crushed to pass a sieve of ⅜-inch mesh, and then heated in the primary retort to 480° C. This retort is horizontal and externally heated. It is about 18 feet long, and is built up of blocks of carborundum, fitted with tongued and grooved asbestos-packed expansion joints. The time of carbonisation is from two to three hours. The retort is of inverted heart shaped cross-section, as shown in Fig. 37, and the material is continuously stirred and moved along the retort by paddles attached to two longitudinal shafts. The paddles have a radius of 2¼ feet, and the shafts rotate one revolution in eight minutes. The end walls are made of firebrick. The retorts are kept half full of material, which is discharged through a water seal, and is then a soft, friable mass. The throughput per retort, if continuously worked, is about 36 tons per day.

The material, on leaving the primary retort, contains from 7–10 per cent of volatile matter. It is then made into ovoid briquettes, the pitch obtained from oils distilled in the plant being used as a binder. The briquettes contain about 15 per cent of volatile matter, and are fed into the externally heated secondary retort. The secondary retort, shown in longitudinal cross-section in Fig. 38, is about 12 feet wide, 21 feet long, and 26 feet high at the discharge end. Each retort is rectangular in cross-section, and contains six carbonising chambers, in two tiers of three each. Being inclined, the briquettes pass through the chambers by means of gravity, and are afterwards quenched in water. The temperature of carbonisation is about 1100° C., and the time taken is six hours. The percentage of volatile matter in the finished briquettes is only 3–4 per cent. The usual by-product plant is installed, and the tar oils and ammonia are recovered from the
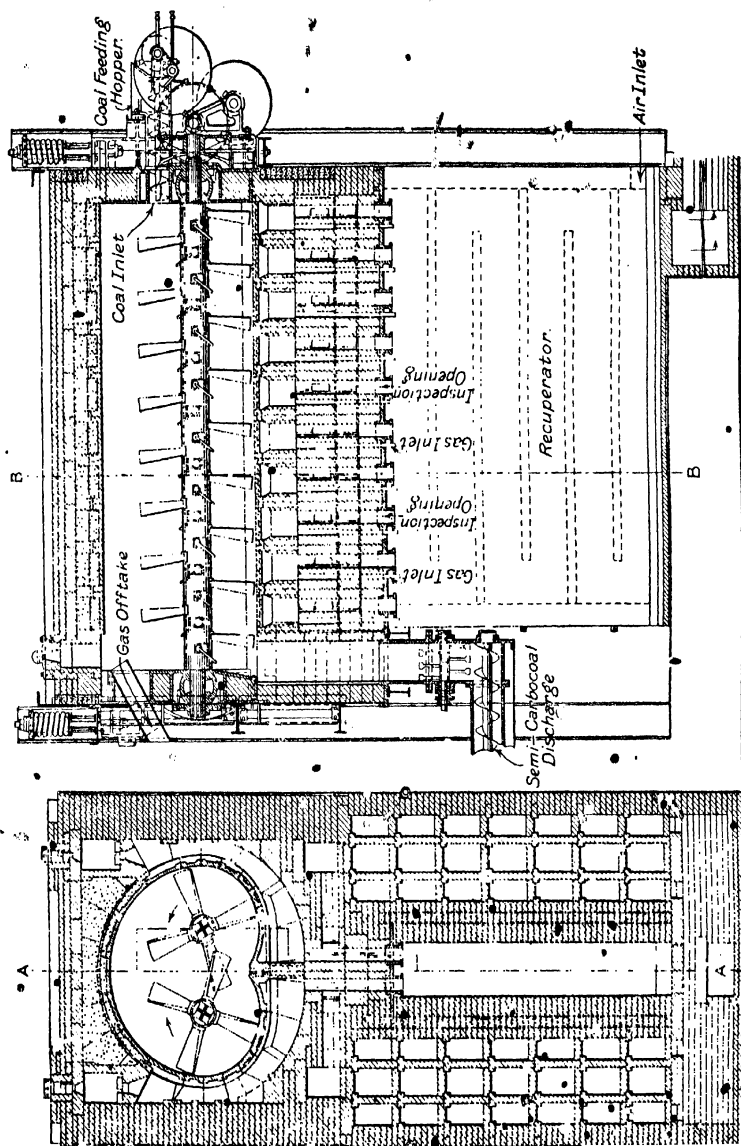
FIG. 37.—CARBOCOAL PROCESS. (Sectional view of Primary Retort.)

gaseous products of combustion. Most of the stripped gas is used in heating the primary and secondary retorts, but there is a surplus which is available for other purposes. The gas burners for both retorts are placed in flues and a recuperative system of pre-heating the air for combustion is employed.
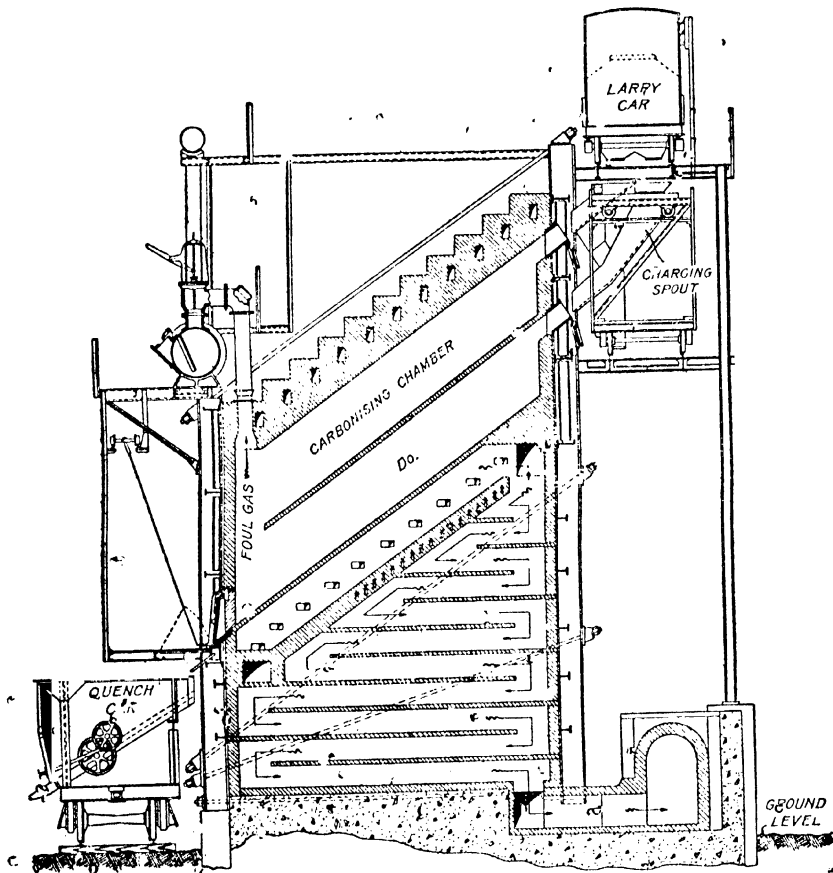
FIG. 38.—CARBOCOAL PROCESS. (Sectional view of Secondary Retort.)

As the pitch has also been decomposed in the second carbonisation, the fuel is quite smokeless. It is very compact and hard, and is thus suitable for transport. It is, however, difficult to ignite. The latter would be a serious disadvantage in a proposed domestic fuel for this country ; but, as carbocoal is in competition with

anthracite in the western states of America, it is quite adapted to the method of burning there adopted. On the other hand, on account of the restrictive legislation, the bituminous coal from which carbocoal may be made cannot be generally employed until it has been pre-treated.

THE EVERARD DAVIES PROCESS

The scheme of operation of this process differs from the other proposals for low temperature carbonisation in certain definite particulars. In the first place, it is claimed that the limits of 10–15 per cent for the volatile content of the coke are superior to the generally accepted limits of 6–12 per cent. The reasons assigned for this are : (a) smokelesness with the higher and lower volatile contents is equal ; (b) the coke with higher volatile content has also a higher calorific value ; (c) there is a saving in the heat required for carbonisation by leaving the extra volatile content in the coke ; and (d) the facility for forming a transportable material is equal to that of a less volatile coke by the control and variation of the retorting factors. It is rightly pointed out, however, that if the higher limit of volatile matter be left in the coke, the volatile content of the original coal should not fall below 30 per cent, or otherwise there is not a sufficient margin for profitable by-product yields. In the second place, the temperature aimed at in this carbonising process is 700–800° C. instead of the lower limits generally adopted by other inventors.

The use of a higher temperature of course reduces the time of carbonisation, and it is claimed that the disadvantages experienced when a higher temperature is used in other low temperature processes are removed in this process by the method of heating employed. In this system, both internal and external heating are proposed. About 25 per cent of the heating gas is passed through the retort, and it is claimed that thereby the time of carbonisation is reduced, so that a given size of retort has a larger throughput than if external heating only were employed, while the small volume of gas employed for internal heating does not reduce the calorific value of the gaseous yield to such an extent as to make the gas unsuitable for town gas. The inventors state that it is their intention to work the retort at either high, medium or low temperature, according to the special circumstances of any installation.

The Everard Davies installation consists of a pair of vertical retorts. These retorts have between them a central vertical chamber connected to the exhauster main, into which pass the gases and volatile products. The retorts are not heated externally throughout their circumference, but only on that part remote from the central chamber. It is claimed that, with this arrangement, the " cracking " of the volatile products of carbonisation is reduced to a minimum. The combustion

chambers are arranged in three or four zones, ranging from 400° C. to about 800° C., so that the material is progressively heated as it passes through the retort under the action of gravity. As previously stated, about 25 per cent of the heating gases passes through perforations in the retort, and mixes with the charge during carbonisation.

'THE FISHER PROCESS

The patented process of coal carbonisation adopted by the Midland Coal Products, Ltd., and worked out by Mr. A. Fisher, B.Sc., A.I.C., is the outcome of investigations begun with the set purpose of finding an outlet for a definite class of poor quality coal. It may be remarked in passing that this attitude towards the problem is very sound, since there are great possibilities of attendant success should colliery owners, or those connected with collieries, attempt its solution in dealing with certain definite types of coal in which they are interested, even should 'the process or retort evolved prove unsuitable for a wider range of material.

The material in which the Midland Coal Products, Ltd., is chiefly interested is non-coking small slack, which is, in normal times, mainly a drug on the market. A typical coal of this type is Top Hard Half-inch Slack, of which the analysis of an average sample is as follows :—

	Per cent.
Ash	31·51
Volatile Matter	24·20
Fixed Carbon	44·29
Sulphur	1·39

Calorific Value, 9800 B.Th.U. per lb.

Naturally the first objective in dealing with material of this nature, in order to produce a final solid product of value, is the employment of an efficient washing process. Economic and effective washing of slacks was not an easy matter until quite recently ; but the Rheolaveur process of coal-washing solves this initial difficulty with satisfaction, both from a technical and commercial point of view. It is stated that the ash content of the raw material of about 30 per cent can now be easily reduced commercially to a maximum ash content of 4½ per cent at a cost not exceeding 5d. per ton, washed.

The raw slack shows on grading and washing tests the following percentages, thus showing the difficulties to be contended with in the washing process :—

GRADING AND WASHING TABLE

SAMPLE OF TOP HARD HALF-INCH SLACK

	Floatings on 1·35 Sp. Gr. Solution.	Sinkings floating on 1·4 Sp. Gr. Solution.	Sinkings floating on 1·5 Sp. Gr. Solution.	Sinkings floating on 1·6 Sp. Gr. Solution.	Sinkings on 1·6 Sp. Gr. Solution.
½ inch to ⅛ inch 69·6% Ash, 22·0% V.M., 27·57% F.C., 50·43%	57% 1·95% Ash	4·5% 7·6% Ash	2·15% 12·9% Ash	2·0% 28·4% Ash	34% 72·3% Ash
⅛ inch to 1 mm. 14·3% Ash, 29·65% V.M., 24·90% F.C., 45·45%	51% 1·8% Ash	3·7% 6·7% Ash	3·9% 12·2% Ash	2·1% 25·4% Ash	39% 71·4% Ash
1 mm. to 0 16·1% Ash, 35·39% V.M., 20·38% F.C., 44·23%	36·4% 2·2% Ash	4·31% 6·4% Ash	5·46% 9·5% Ash	1·13% 20·6% Ash	55% 53% Ash
		Tailings Ash 73·50%			

The next problem was to put this fine washed coal into suitable form to find a market other than for boiler-firing purposes, for which, of course, such a clean material has an immediate outlet. Straight carbonisation in the state of washed slack was in any case a useless procedure to attempt, as the coal was of non-coking character, and the carbonised product would consist of nothing but breeze, for which no satisfactory market existed.

Briquetting of such breeze by the addition of a binder to the carbonised fuel, by which means volatile matter would again be added to the carbonised material, seemed an unscientific procedure. Complete gasification was a possible outlet,

but here again the material was too small to be of general use for this purpose, and this, combined with a low fusibility of ash, restricted the market.

In order to get a satisfactory solid product in a marketable condition, lump material was essential. This has been attained by simple briquetting before carbonisation.

Ordinary briquetting with normal pressures, entailing normal disintegration of the slack, was considered essential from a commercial point of view for many reasons, but it was obvious that with a binder of the nature of pitch introduced into the fuel, it would be impracticable to carry out satisfactorily any method of carbonisation by external heat application, owing to the tendency which the binder would have to flow out of the material at a temperature lower than that of the incipient fusion of the coal substance. Internal heating was experimented with and satisfactory results obtained. It was found that by grinding the washed slack to a grade of fineness to pass an ordinary ten mesh before briquetting, the incorporation of the binding agent assisted the coking of this particular coal.

Here it is to be remembered that the Top Hard seam is a banded seam of dull and bright coal with a quantity of black shale, but a higher concentration of the bright coal, having a higher coking index than the dull hard bands, is found in the slack, and by grinding and mixing, the non-coking particles are found after carbonisation to be intimately mixed with the higher coking particles of the bright material, and a uniform hard coke is produced. By this means a natural blend of coking and non-coking coal is obtained at once, such as has been aimed at by scientific blending of coking and non-coking coals artificially for several processes carrying out low temperature carbonisation.

The briquettes have a maximum weight of 5 ounces and are made ovoidal or spheroidal in shape, with the definite object of ensuring regular interstices throughout the mass, not only for the penetration of the blast introduced at the base of the retort, but also for the full utilisation of the sensible heat of the liberated gases by uniform distribution throughout the mass of the oncoming charge.

The actual process of carbonisation is thus much on the lines of pressure gas producer practice, with the exception that the air-steam mixture is fed in horizontally from the periphery of the retort base. The retorts themselves resemble miniature blast furnaces in shape, tapering upwards and downwards from a maximum diameter of 5 feet 6 inches at the widest part, with cooling arrangements, at the bottom.

The ovoids are fed in at the top to maintain a bed of 12 to 14 feet depth, and gravitate through the retort without any internal mechanism. It is found that, by proper control of blast conditions, considerable variation in throughput can be obtained without changing the quality of the product. A maximum throughput of 28 cwt. per hour per retort is obtainable.

The feared disruption of the briquettes in passing through the retorts does not take place to any marked degree ; and under normal conditions a 60 per cent discharge of whole briquettes is obtainable, 80 per cent of the remainder being halves and quarters, with an almost negligible proportion of dust.

Naturally, with a process of this description, good briquetting is of vital importance.

The maximum temperature obtained is in the region of 1000° C. in the combustion zone, and the volatile content of the hard carbonised product is between 1 and 2 per cent.

Contrary to general anticipation, this material is a free-burning fuel.

There is a tendency for the high heats obtained by forced draught conditions to fuse the ash into clinker, but this is due to the inherent quality of the fixed ash in the coal itself, which has a low fusion temperature. Other coals with a different ash do not exhibit this quality to the same degree. A number of coals, particularly Staffordshire coals, have been treated, and it is found that the process is applicable to a large variety of coals with a low coking index.

The Midland Coal Products, Ltd., have now erected a battery of four retorts with a normal capacity of 100 tons throughput per day, and are obtaining full commercial data on the process.

The particular features of the process of importance from the economic point of view are : The low capital cost of plant in relation to the throughput, and the low rate of depreciation of the retorts owing to the small area of brickwork which is subjected to destructive heat.

THE FREEMAN MULTIPLE RETORT

The Freeman Multiple Retort can be used with oil shale or non-caking coals which do not fuse when they are heated. The retort (Fig. 39) consists essentially of a series of cast-iron chambers placed vertically one above the other. The coal enters the top chamber through a gastight charging valve, and falls upon a rotating plate. By means of ploughs it is spread over this plate in a thin layer and is gradually worked to the outer edge, whence it falls on to the bottom of the first chamber.

A series of ploughs attached to the bottom of the rotating plate gradually works the coal into an opening in the centre of the floor of the chamber ; thence it falls upon a rotating plate in the second chamber, and the process is repeated. The coal is therefore very evenly heated up to the maximum temperature. The first chamber is maintained at a temperature approximately 120° C., sufficient to drive off the combined and free moisture in the coal.

The second chamber brings the coal up to the temperature of incipient oil

3'-4"

1'-9"

2'-4"

5'-2"

2'-4"

4'-8"

2'-4"

2'-4"

27'-2"

2'-4"

2'-4"

2'-5"

8'-0"

ALL SECTIONS
1IN. THICK.
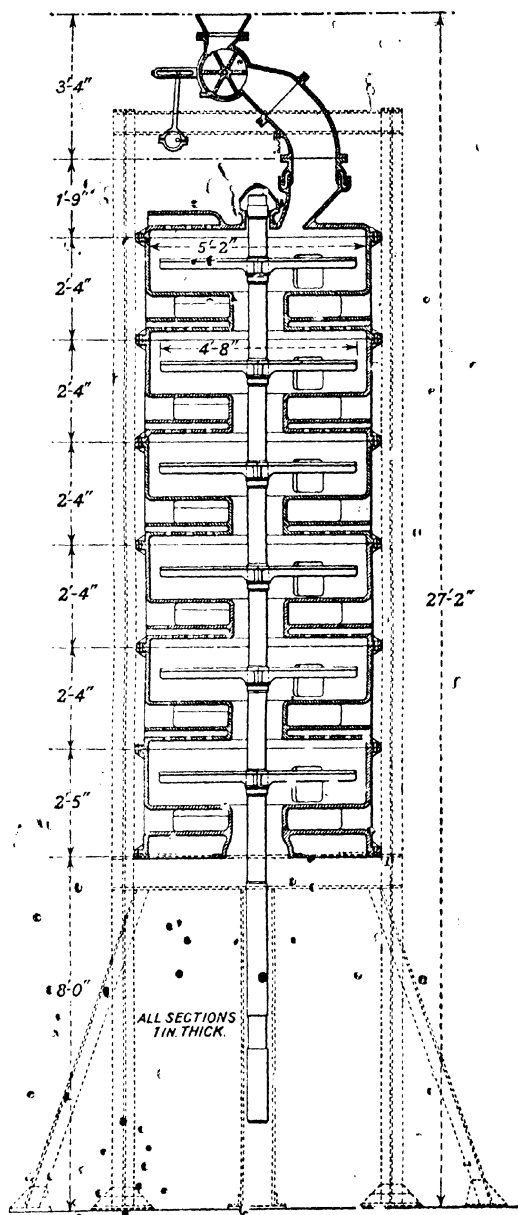
FIG. 39.—FREEMAN MULTIPLE RETORT.

production, while the maximum temperature attained in the fifth or sixth chamber is about 455° C. to 485° C.

The original idea in connexion with this plant was that the coal should be fractionally distilled. An attempt was made to isolate the distillation products given off at each temperature. This, however, has in practice only been partially successful. The first chamber does act practically as a dehydrating chamber, and the products of distillation from the top chamber consist almost entirely of water. The oils and tars from the remaining chambers are not clean-cut fractions, however, and some of the oils from chamber No. 2, for example, are collected in receiver No. 3, and so on. Despite the general lack of fractional distillation, the removal of the water in the first chamber does materially reduce the dehydration necessary of the final oil products.

A plant has been installed at Willesden of a capacity of ten tons per day. The temperature regulation has proved to be exceedingly efficient, and the system has demonstrated its ability to deal successfully with non-fusible coals and oil shales. The system is of use where smokeless fuel in powdered form is required for dust firing or for subsequent briquetting.

THE FUEL RESEARCH BOARD RETORTS

It has been previously pointed out that one of the difficulties in the development of low temperature carbonisation is that laboratory experiments on small quantities of coal do not necessarily give the same proportionate results as those obtained when the material is treated in large quantities on an industrial scale. The determination of the relations between operations on a laboratory and works scale is therefore of some importance, and it was with this underlying idea that the large-scale experimental retort of the Fuel Research Station was designed. The fundamental provisions it was desired to embody were :—

(i) The steady maintenance within the retorts of any desired carbonising temperature up to 600° C.

(ii) The exposure to the desired temperature of coal crushed to various degrees of fineness in layers of any desired thickness, due regard being paid to the effects of heating by radiation, conduction and convection.

(iii) The collection and accurate measurement of the gases and volatile liquids resulting from carbonisation.

(iv) The removal, quenching and weighing of the resulting coke and the study of the influence of fusibility in different types of coal on the physical properties of the coke.

(v) The design and construction of the ovens or chambers in which the carbonising retorts or machines are set so as to ensure uniformity of heating

with the minimum expenditure of fuel and the minimum deterioration of the metal of which the apparatus is constructed.

(vi) Towards the attainment of these ends the complete control and accurate measurement, not only of the fuel gas used to heat the setting, but also of the air supplied for its combustion.

The setting of the retort was designed before the laboratory assay apparatus had reached its final form, but the importance of studying the behaviour of the
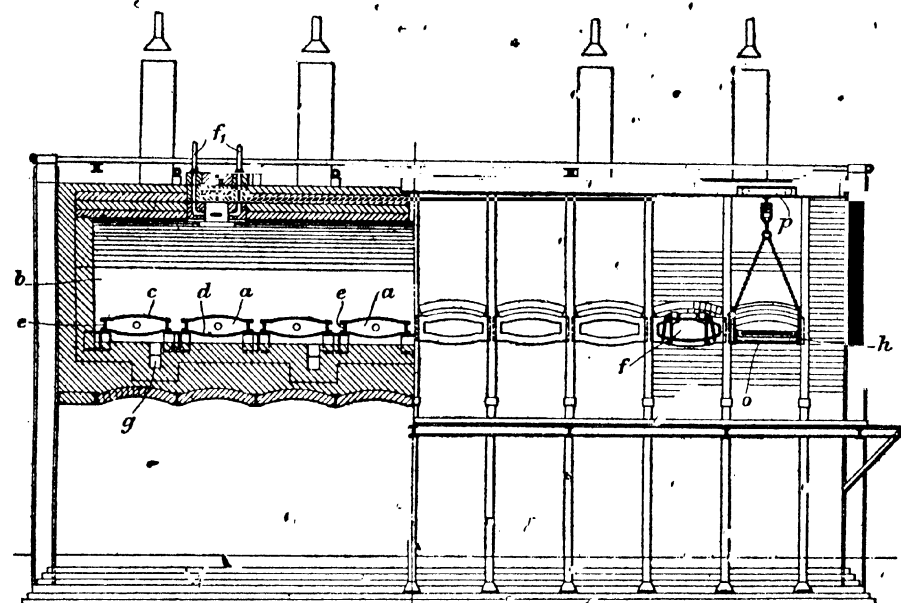
FIG. 40.—H.M. FUEL RESEARCH STATION. HORIZONTAL STEEL RETORTS FOR EXPERIMENTAL WORK ON LOW TEMPERATURE CARBONISATION.

Sectional elevation of Bench.

a. Steel retort.   *b.* Combustion chamber.   *c.* Cambered steel plate (top).   *d.* Cambered steel plate (bottom).   *e.* Channel iron side (5" deep).   *f.* Self-sealing door.   *f₁.* Nostril blocks for fuel gas and air.   *g.* Waste gas flue.   *h.* Carbonising trays.   *p.* Overhead traveller.   *o.* Movable cradle.

carbonisation of coal at different temperatures and of preparing accurate balance sheets was kept in view throughout in both cases.

A full description of the horizontal steel retorts erected at H.M. Fuel Research Station is given in the Report of the Fuel Research Board,[1] but a sectional elevation is shown in Fig. 40. Briefly, the battery consists of nine horizontal retorts, each

[1] Fuel Research Board. Report for the Years 1920, 1921; Second Section: "Low Temperature Carbonisation." (H.M. Stationery Office.)

9 feet long and 2½ feet wide. Each retort consists of top and bottom plates of ⅜-inch mild steel, slightly cambered, connected by 5-inch standard section channels. With this cross-section it has been found that t ie retorts have successfully resisted the tendency to distortion due to their exposure to a temperature of 600° C. during a lengthy period. The retorts are closed at the front, and by self-sealing doors through which are inserted the trays in which the coal for carbonisation is spread. Two trays per retort are used, each tray being the full width of the retort and 3 inches deep. Steel strips across the trays divide the material into blocks of 3-inch cube. The gas off-take is at the back of the retort and leads to a common collecting main, but the off-take is automatically closed when the retort door is opened. The vapours are led as usual through condensers, scrubbers, etc., for the collection of the oil, etc., and the gas is collected in a gas-holder.

The heating chamber for the battery is 30 feet long, 8 feet wide, and on the average 4 feet high. It has a capacity of about 860 cubic feet. The gas and air are supplied through three nostril blocks in the roof. The combustion of the gas is completed under the crown of the arch, which is maintained at a temperature of over 600° C. and radiates its heat to the upper surfaces of the retorts. The products of combustion pass the retorts on their way to the flues which lead from the bottom of the chamber.

These retorts have proved extremely useful for the purpose for which they were designed. The yields and qualities of the products of the carbonisation of various types of coal have been ascertained with great accuracy, and valuable knowledge of the maintenance of retorts of this description has been obtained. The most serious disadvantage from a commercial standpoint has been found to be the relatively heavy labour charges involved on account of the carbonising trays being put in and taken out of the retort by hand. Even under such adverse conditions, however, the retort at one state of the market was practically self-supporting.

In view of the experience gained by the working of these retorts, a further carbonising machine was devised and patented,[1] one of the patent drawings of which is shown in Fig. 41. It will be seen that the carbonising trays rest on an elevator which can be moved vertically in a gastight chamber. The trays can in turn be moved opposite an opening in the retort through which the tray is drawn to an outside chamber. It is then discharged of its coke, reloaded with coal and returned to its shelf in the elevator. This retort has been in operation for some months, and very satisfactory coke has been produced. On account of the reduction in the amount of heat transferred to the coal by radiation, an increased time of carbonisation has been required over that necessary in the earlier retort, and the mechanical difficulties caused by the tendency of parts to get out of register owing to "creep" of the metal at the temperatures used have likewise reduced the

[1] Pat. Spec. 178,934 (1922).
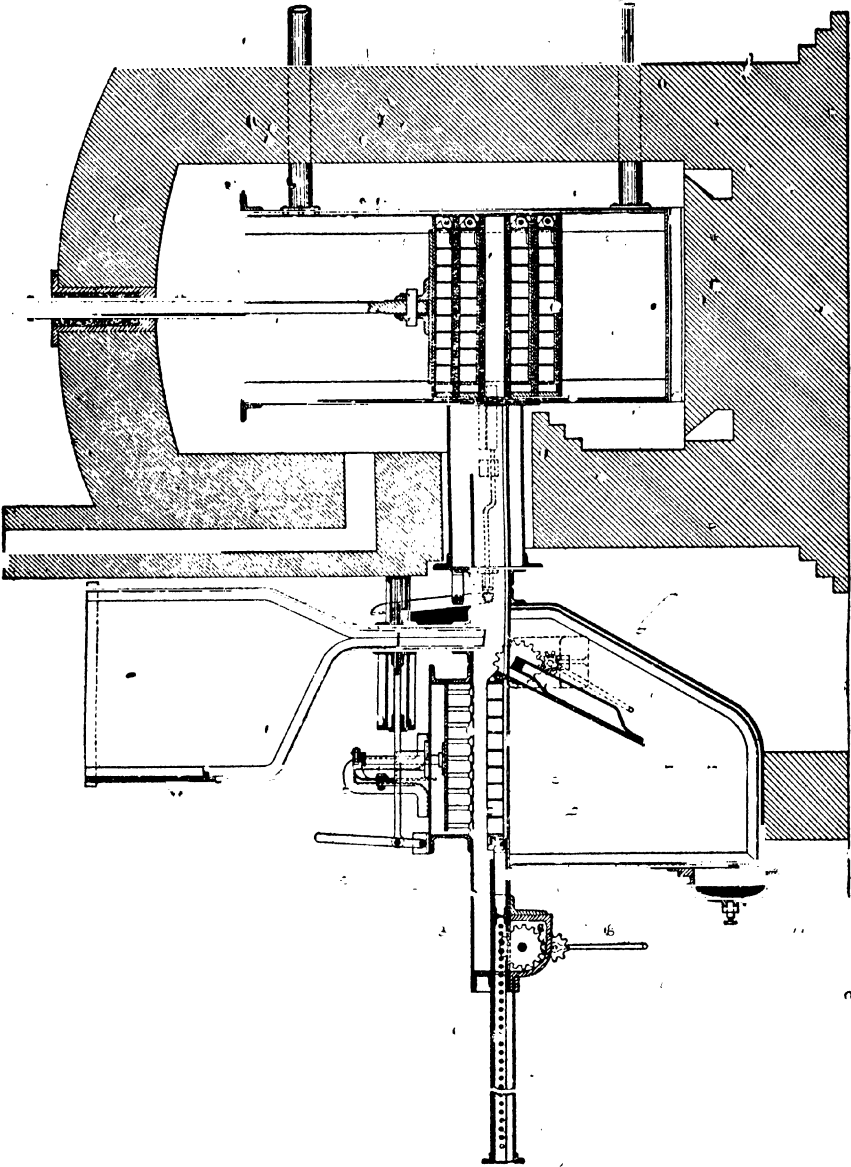
FIG. 41.—H.M. FULL RESEARCH STATION. EXPERIMENTAL CARBONISING MACHINE.

advantages which would otherwise have been obtained. The experiments upon this type of apparatus have accordingly been dropped for the time being, as it seemed that a more promising method of low temperature carbonisation was opening out in another direction, and the time of the staff and the facilities of the station might more profitably be used in developing the new scheme. (See p. 261.)

THE FUSION PATENT LOW TEMPERATURE RETORT

The Fusion Patent Low Temperature Retort is made in two types, viz. double tube type and single tube type. In the former, the material passes down the centre of the inner tube, where it falls into the outer tube. It then passes down the

LONGITUDINAL SECTION

CROSS SECTION
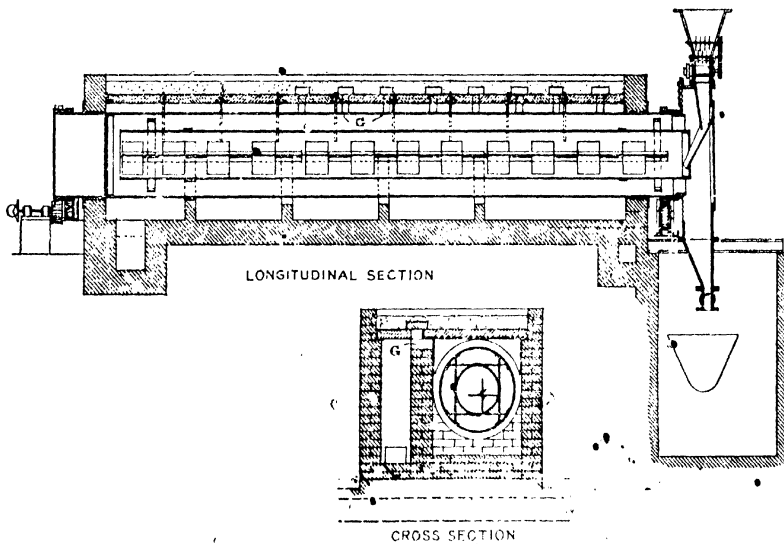
FIG. 42.—FUSION PATENT RETORT.

annular space between the inner and outer tubes, and leaves the outer tube at the same end at which the material is fed to the retort. In the single tube type the material is fed into the tube at one end and discharged at the other. In each case the axis of the tube is horizontal. In the double tube type, the tubes are concentric, and in the design illustrated (Fig. 42), the outer is about 30 inches in diameter and the inner about 18 inches in diameter. The retort is constructed in standard sizes from 2 feet 6 inches to 4 feet in diameter, and from 25 feet to 100 feet in length. The capacities of these retorts vary from 8 tons to 100 tons of coal per 24 hours.

It is stated that for a speed of 5–7 revs. per minute, 1 B.H.P. is required to drive the 5-ton unit.

Within the inner tube is a loose " star " or " cross " breaker, whose diameter is about two-thirds that of the internal diameter of the inner tube. As the retort rotates the breaker is carried with it until a point is reached when the breaker falls over, and one point or edge of the star or cross comes in contact with the bed of the material under treatment, giving it a glancing blow with a chipping effect. A similar type of breaker may be employed in the annular space between the tubes, or alternately the inner tube may be provided with external ribs and act itself as a loose breaker in the outer tube.

The material, which is preferably crushed to pass through a ⅜-inch sieve, is fed into the inner tube at one end and leaves the outer tube at the same end, after
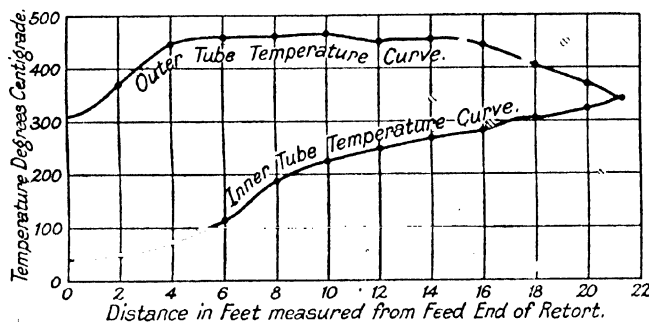
FIG. 43.—FUSION PATENT RETORT.

Typical Temperature Gradient.

traversing first the inner and then the outer tube. The material is not mechanically propelled along the tubes, but moves along through finding its angle of repose as the tubes rotate. There must, of course, be a difference in level of the material at inlet and outlet for motion to take place. This difference in level, or " head," varies with the rate at which the material is treated, with the speed of the rotation of the retort and with other factors, but it is stated that the head required in a single-tube retort is about 2 inches.

The heating gases circulate around the outer tube, and hence the inner tube is only indirectly heated. The temperature gradient of the material in the retort therefore rises very gradually ; and, as given by Capt. U. J. Goodwin,[1] is shown in Fig. 43 for a typical run. By manipulating the dampers G (Fig. 42) the temperature gradient in the outer tube may be regulated to suit different materials,

[1] Goodwin, C. J. " The Fusion Patent Low Temperature Retort." *Trans. S. Wales Inst. of Eng.*, April, 1922.

rising either gradually or quickly to a maximum, or rising in defined stages if it be expected that a strongly marked exothermic action will take place in the material. The evolved gases and vapours in each tube remain separate until they mix on exit.

A special mechanical feature of the retort is the design of stuffing box at the exit end of the tubes. The method of making the joint is illustrated in Fig. 44. The joint is made between an asbestos compound ring and a steel ring. The asbestos compound ring is secured to the revolving tube, while the steel ring is carried by a diaphragm which is corrugated in order to increase its flexibility. Steel springs are employed to keep the rubbing surfaces in contact.
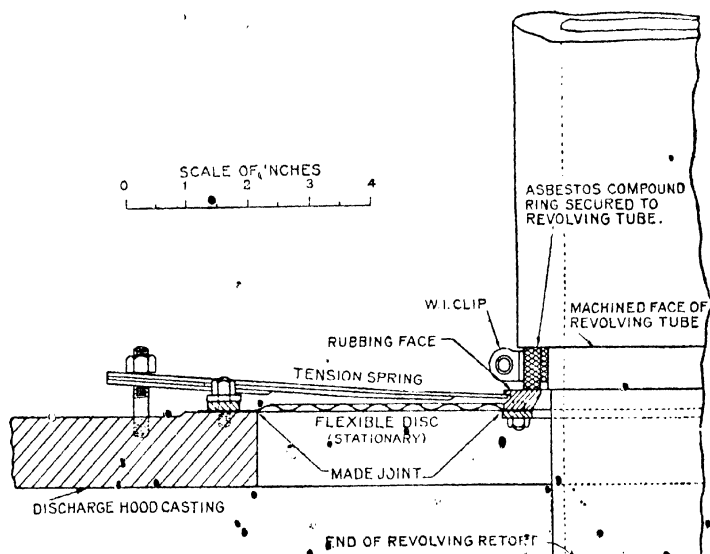
FIG. 44.—FUSION PATENT RETORT. Detail of Flexible Joint.

The loose breaker differentiates this retort from other somewhat similar devices. The outstanding claim made is that the breaker not only keeps the heating surfaces free from dirt and scale, but that it also prevents the caking of the material itself. The power required to actuate the breaker is very small; and, as a scraping action takes place each time the breaker falls over, there is no danger of the scraper breaking or failing to function. It is stated that the throughput of a Fusion retort of the dimensions described, is from 3½ to 5 tons per 24 hours.

An experimental retort of the above type, though without the inner tube, has been constructed by the Fusion Corporation, Ltd., at their works at Middlewich, Cheshire. Its throughput is 5 tons per day.

THE MACLAURIN PROCESS

The earlier developments of the Maclaurin process have already been outlined on page 200. The most recent development arises from the decision of the Glasgow Corporation to install a battery of five units of this type of retort at Dalmarnock. Two objectives are aimed at : Firstly, the production of smokeless fuel, and secondly, to prove whether or not the gas obtained by the process can be economically used for the firing of the boilers in the generating station.

Fig. 45 shows one of the latest types of the Maclaurin retort. It has a square
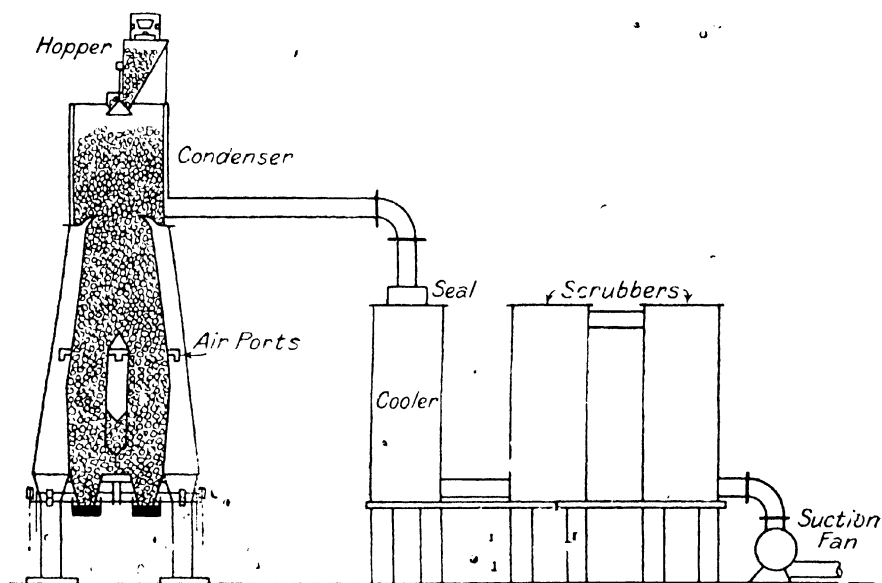
FIG. 45.—MACLAURIN CARBONISING PLANT.

cross-section, its maximum internal width being 8 feet and its overall height about 45 feet. The retort is raised above ground level so that tubs can be run beneath to facilitate discharging. For the purposes of securing uniform distribution, the air blast enters through a large number of narrow ports in the opposite side-walls, and also by similar ports in a dividing wall which is carried across the plant at the same level. The zone below the entry of the air is the cooling zone. Steam is injected at the discharging doors, and, in cooling the coke before discharge, becomes heated and partly decomposed into water gas. Passing upwards, it mixes with the producer gas formed at the combustion zone. The cylindrical tank at the top

of the retort, which acts as a condenser, has a diameter of 8 feet and a height of about 10 feet. The bottom of this tank is dished upwards, so as to provide a well for the collection of the oil and water. An open internal cylinder dips into this well, leaving an annular space of about 2 inches for the passage of the gas.

The plant is continuous in operation; and a retort of the dimensions given contains about 30 tons of fuel. It may be run either for complete gasification of the fuel or for the production of smokeless fuel, the only adjustment necessary in changing over consisting in the regulation of the air blast and gas outlet valves. It is stated that the gas leaves the plant at a temperature of from 60° C. to 80° C., so that the heat losses involved in the process are very small.

THE MARSHALL–EASTON PROCESS

The Marshall–Easton retort, a suggested arrangement of which is illustrated in Fig. 46, is of the vertical type, heated internally by the sensible heat of water gas and heated externally by the ignition of " blow gas " in the surrounding chequer-work. The retort is made of silicon cast iron, $1\frac{1}{4}$ inches thick, and consists of two vertical cylinders with a common segment, so that the cross-section is in the form of the figure 8. The interior of the retort contains two screw conveyors, one in each cylinder. The screws are so disposed that the projecting portion of one is opposite the recess of the other in the common segment of the retort. The movement of the screws is synchronised, the speed of each being from six to ten revolutions per hour. The descent of the charge through the retort is controlled by the movement of the screws, and the claim is made that, as the blades of the screws segmentally overlap, highly bituminous or sticky coal cannot hold itself up in the retort, as it is continually being wiped forwards and downwards. It should be noted that the claim that the screws act as scrapers is not made by the inventors. The charge progresses through the retort in segmental rings about 6 inches deep and about $4\frac{1}{2}$ inches thick. Thus the external heat of carbonisation has to pass only through the thickness of the retort and $4\frac{1}{2}$ inches of the charge, and it is claimed that the time of carbonisation is from three to four hours. A full description of the retort and process is given by Mr. F. D. Marshall in a recent number of the *Gas Journal*.

THE PEHRSON PROCESS

The Pehrson rotary furnace system is designed to separate the products which come off when coal is heated up to 250° C. (or higher with some coals) from those which are evolved between that temperature and 560° C. It has already been shown (Chap. VII) that the former consist mainly, if not entirely, of carbon-

---

[1] Marshall, F. D. "Low Temperature Carbonisation by the Medium of Water Gas: The Marshall–Easton Process." *Gas Journal*, June 13, 1923.

dioxide and the water of constitution, whilst the oils and hydrocarbon gases are evolved during the latter stage. In the higher temperature stage internal heating is effected by recirculating a proportion of the make gases after their temperature has been raised in a separately fired gas-heater. In the lower temperature stage the internal heating is effected by means of the flue gases from the gas-heater.

The process is intended to treat small coal or duff. The main operations are carried out in rotary retorts which are intercommunicating but which allow the products of distillation mixed with the heating gas to be taken off separately from each furnace.

The retorting apparatus consists of a drying tray and two rotary furnaces. The fine coal first passes over a drying tray which consists of a circular table under which the hot gases from No. 1 furnace are conducted. It is then fed into No. 1 furnace, where it is treated by hot air or waste gases at a temperature below that at which combustible volatile matter begins to be liberated to any extent. The distillation products from this furnace being valueless are discarded after passing under the drying tray, but a portion is circulated through a heater and reintroduced into the furnace. No. 1 furnace is at a higher level than No. 2 furnace in order to facilitate the transference of the charge. The material enters No. 2 furnace at a temperature of from 250° C. to 300° C. and then gravitates gradually towards a zone of heat which at the discharge end is kept at about 560° C.

The rotary furnaces consist of deep drums, each of which has tubular extensions or necks at both ends to facilitate charging and discharging. Integral with the drums are a number of conduits through which the hot gases are introduced into the respective charges. The method of conveying the hot gases to the charge is by means of valves so designed that the gas is forced into the charge through a number of tubes inserted in a circle round the periphery of the furnace and maintained through to the opposite side. These tubes have longitudinal openings at given intervals, and it is stated that they are so constructed that the orifices cannot be choked by the charge.

The valves are so designed that only those tubes which are below the charge of coal at a given depth are in service. It is affirmed that this device gives perfect control since the gas is always passed through fuel of determined depth, which being in constant motion can be maintained at a very uniform temperature whilst the risk of condensation of oils within the charge is reduced to a minimum.

THE PURE COAL BRIQUETTE PROCESS

The Pure Coal Briquette process which has been developed by Mr. E. R. Sutcliffe and Mr. E. C. Evans depends for its success more upon the pre-treatment of the material than upon the type of retort used. The authors indeed state that their

process can be successfully carried out in existing gas retort or coke oven plants, although a special type of retort has also been designed when a smokeless domestic fuel is required.

The coal is first washed and dried, and then ground to the size determined by the characteristics desired in the resulting coke. The size usually recommended
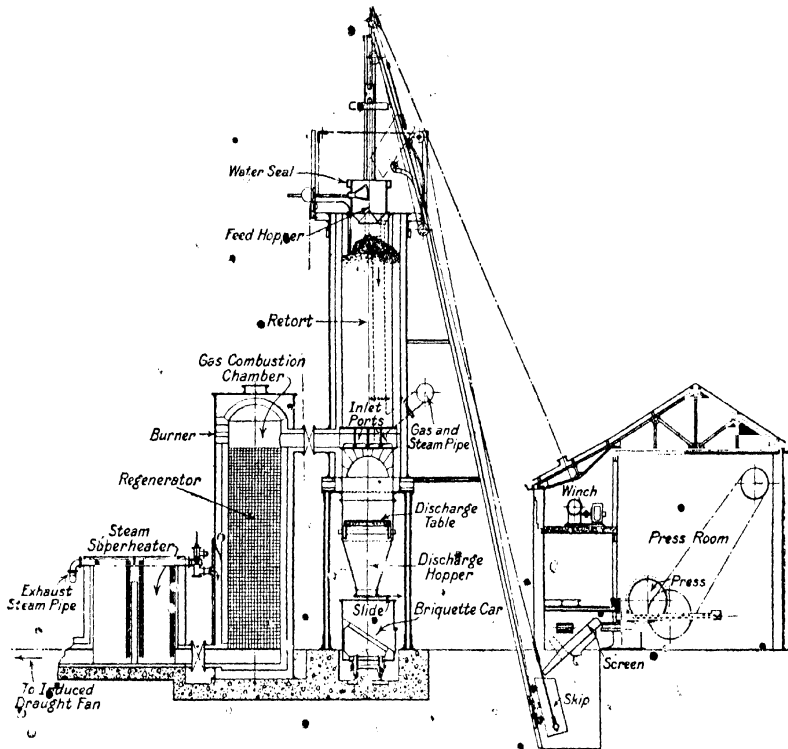
FIG. 47.—PURE COAL BRIQUETTE PROCESS.

is 30 mesh. The coal is then mixed with coke breeze, non-caking coal or pre-heated coal in the requisite proportions to render the mixture non-expanding. It is stated that, with most British coals, good results can be obtained by the addition of 20 per cent to 25 per cent of coke breeze. The mixture is then briquetted without a binder, a pressure of eight to ten tons per square inch being applied in a specially designed ovoid press. The briquettes thus produced are hard and dense, and not only pass through the retort without fracture, but result in a fuel which does not

readily disintegrate during transit. The inventors state that a briquetted material of this description may be advantageously used in gasworks and coking ovens. The coke is not only free-burning and smokeless, but is hard and dense. It is claimed to be of better quality than the cokes obtained from existing processes.

In order to obtain the most suitable fuel for domestic purposes, the material is subjected to a gradually increasing temperature which may rise to 1000° C. The resulting fuel has a low volatile content, but it is claimed that on account of its structure the coke is as easy to ignite and as free-burning as a coke with a volatile content of 10 per cent. The use of this high temperature ensures a high yield of gas and ammonium sulphate. In addition, since the coal is gradually heated to its maximum, the tars are expelled at low temperatures and have the same characteristics as ordinary low temperature tars.

The type of retort employed is shown in Fig. 47. The retort consists of a vertical cylindrical chamber lined with firebrick and lagged with insulating material. It is heated internally by means of steam superheated in preheaters and regenerators to the required temperature. The retort is fitted with two regenerators, which are used alternately in the usual way, one being heated while the other is giving up its heat to the steam used as the heating medium. The regenerators are heated by part of the surplus gas being burned at the bottom, the products of combustion passing through the chequerwork bricks.

THE SCOTT-MONCRIEFF PROCESS

In the Scott-Moncrieff process, of which a detailed description has not yet appeared in the technical press, though particulars are given in his Patent Specification,[1] the material is placed in a series of trays, which are superimposed one on the other in a cradle or support. The trays are fitted with circular bosses or distance pieces, which engage by male and female projections, and leave a free passage between the trays for the withdrawal of the gaseous or vapour products of carbonisation. Means are provided for engaging the trays to the cradle, so that the trays of the series are successively introduced or withdrawn from the lower end of the latter. The cradle is rigidly attached to the lid of the retort, and can be conveniently introduced into and removed from the retort by means of an overhead traveller or trolley. After the material has been carbonised, the trays are removed from the retort and placed in a cooling chamber of the same size as the retort. The circular groove on its upper rim corresponds to the tongue on the lid of the retort, so that a tight junction is readily effected. When the material has been cooled, the cradle is taken to the unloading platform, and the trays disengaged.

Hydraulic power is suggested as a convenient method of raising and lowering

[1] Scott-Moncrieff, W. D. Pat. Spec. No. 182,888 (1922).

the cradle for this purpose. When the trays have been unloaded, they are taken to a loading platform, where the correct amount of fuel is introduced into each. The operations of loading the cradle and carbonising the material are then repeated.

It will be seen that the principle involved in this process is very similar to that incorporated in the Fuel Research Board apparatus described on p. 221. Mr. Scott-Moncrieff's further experiments will be followed with great interest.

THE "SENSIBLE HEAT" (NIELSEN) PROCESS

In the "Sensible Heat" (Nielsen) process the retort is of the inclined rotary type, and is internally heated with hot producer gas. After carrying out a considerable amount of experimental work in a small plant erected at Chatterley, Messrs. Bryan Laing and Harald Nielsen, the owners of the patent rights, have recently installed a large plant at the works of the Carbon Products Company in India. This retort, which is designed to treat 100 tons of coal per day, is illustrated in Fig. 48. It consists of a steel shell, which is about 90 feet long. Over a length of 40 feet the diameter is 7 feet, and over the remainder of the length the diameter is 9 feet, the larger diameter being at the discharge end. Near the lower end an annular chamber, 14 feet in diameter and 8 feet long, is built round the shell. This chamber is for the purpose of cooling and facilitating the discharge of the material after carbonisation. The shell is lined with firebrick as a protection against the action of heat, and is lagged on the outside in order to reduce radiation losses. The charge is fed into the retort at the upper end, and the inclination and speed of the retort are such that complete carbonisation is effected when the material has reached the lower end of the retort. Separate discharging doors into and from the cooling chamber are fitted and are hand-operated.

The retort is driven through a spur-ring which surrounds the shell at its mid-length. The weight of the retort is carried on rollers by three running bands, which are situated one near each end of the retort and one near the driving band. End thrust is taken by thrust rollers resting against the side of the running band.

The material is carbonised by means of hot producer gas which traverses the full length of the retort in the opposite direction to the movement of the charge. The producer, which is of special design, is placed near the lower end of the retort. The outlet from the producer enters the revolving portion of the retort through a specially designed gland in which cast iron rings rest on metallic surfaces. At the other end of the retort, a fixed cylindrical casing, 3½ feet in diameter, carries both the coal-feeding mechanism and the gas outlet branch. The gland at this end is similar to that at the other. In their design the inventors appear to have solved the difficult problem of keeping a large-diameter stuffing box tight under

high temperatures. The coal is fed into the retort by a screw conveyor working in a trough into which the coal falls from a mechanical measuring device.
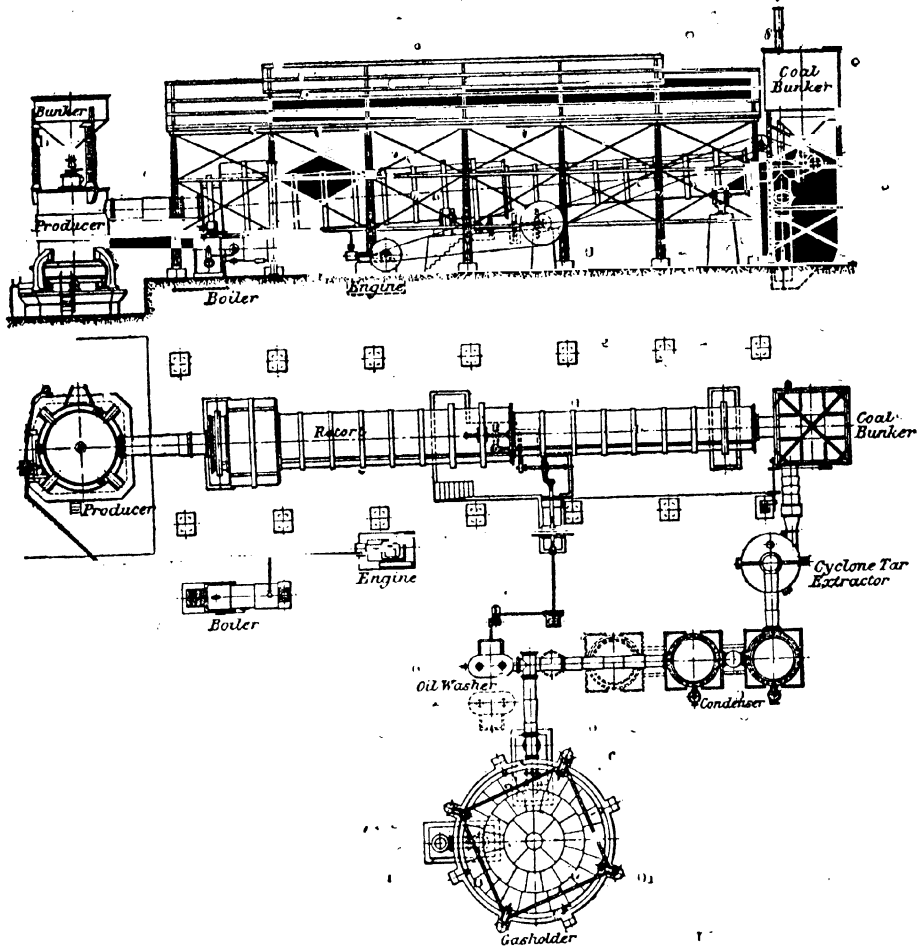
FIG. 48.—" SENSIBLE HEAT " (NIELSEN) PROCESS. PRODUCER GAS SYSTEM.

The hot producer gas in its passage through the retort is enriched with the products of distillation. After leaving the retort it is passed through a tar extractor and condenser for the separation of the oil before it is used or collected in a gas-holder.

A further experimental plant to carbonise 10 tons per day, is to be erected at Silkstone, Yorkshire. In this plant, illustrated diagrammatically in Fig. 49, the inventors intend to use hot producer gas as the heating agent only when the plant is being started up. When in full operation the make gas after being condensed and oil scrubbed will be passed through a superheater placed in front of the rotating retort and then through the retort, so effecting carbonisation by means of its sensible heat. The superheater is so arranged that it can be fired, either by using a portion of the scrubbed make gas, or by producer gas. In this way it is hoped to obtain a gas more nearly approaching the rich gas that can be produced in externally heated retorts, whilst retaining the advantages associated with internal heating.

THE TOZER PROCESS

The main features of the Tozer process have already been described on page 197, and there is little to be added to the account there given. A large plant of 24 retorts with a throughput of 120 tons of coal per day is now in course of erection at Ballengeich, Natal, for the South African Carbide and Products Co., Ltd., and the results obtained from this plant will be awaited with much interest.

A considerable amount of work has recently been done on lignites from various parts of the world, and a first section to treat 100 tons per day of 24 hours is now in course of erection in France, the plant being designed to treat ultimately 500 tons per day.

OTHER PROCESSES

It is not possible to describe all the processes and retorts suggested for the low temperature carbonisation of coal, but a summary of the distinctive features of others not already described is given in Table XLVI. It is thought indeed that a tabular description along these lines of all the processes and retorts already considered would be interesting and instructive, but limitations of space render such duplication in this volume inadvisable.

SUMMARY OF SUGGESTED METHODS OF RETORTING

From the foregoing description of apparatus, it will be seen that inventors have suggested a large number of types of retort. In addition to those which have been described in some detail, there are many others found in the pages of the patent literature of this and other countries. Most of these are likely to remain there, some as records of misplaced ingenuity, and others as tombstones of unscientific skill. In common with many other inventions, the problem has usually been approached from too limited an angle. For the attainment of success, the help of the engineer, physicist and chemist is required. Co-operation is not only essential
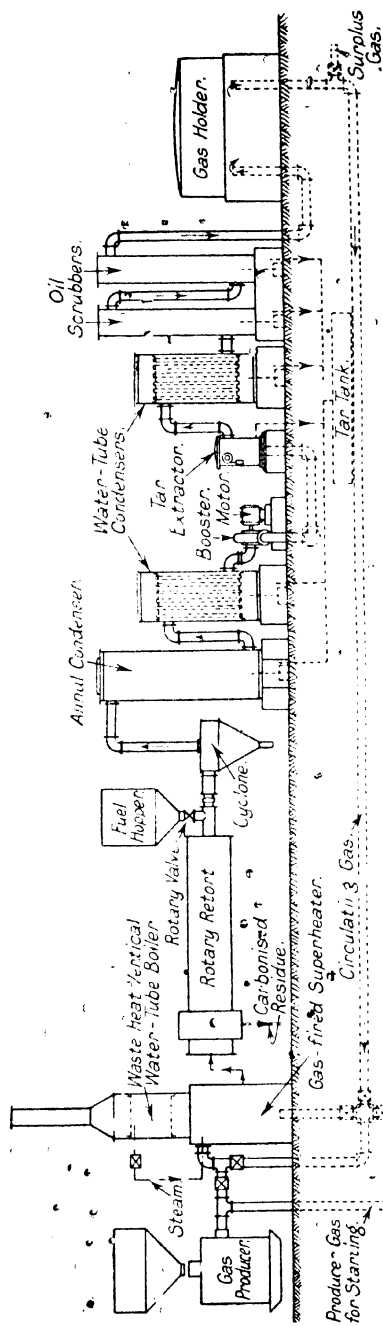
FIG. 49.—"SENSIBLE HEAT" (NIELSEN) PROCESS. SUPERHEATED CIRCULATING GAS SYSTEM.

TABLE XLVI

MAIN FEATURES OF OTHER PROCESSES

	Fellner-Zeigler	Thyssen	Raffloer	Sumniers	Burney	Lamplough	Turner
1 Country	Germany	Germany	Germany	America	Great Britain	Great Britain	Great Britain
2 Retort :— Direction of axis of retort	Inclined 1 in 20	Horizontal	Horizontal	Horizontal	Horizontal	Superimposed horizontal retorts of small diameter connected by headers	Vertical
3 Movement of retort	Rotary	Rotary	Rotary. Inter-mittent motion	None	None	None	None
4 Approximate size of retort	65½ ft. long × 8½ ft. diameter	76½ ft. overall, 5¾ ft. high	About 35 ft. × 8 ft. diameter	40 ft. long × 17 in. wide × 27 in. high			
5 Material of retort	Steel retort. Brickwork setting	Steel retort. Brickwork setting	Steel retort. Brickwork setting	Firebrick	Cast iron	Cast iron. Brickwork setting	Iron
6 Charge :— Size of material	About 1 in.		Pulverised		About 1 in.		
7 Movement of charge— (1) Continuous or Intermittent (2) How effected	(1) Continuous (2) Rotation and inclination of retort	(1) Continuous (2) Spiral ribs within retort	(1) Continuous (2) Material only in annular chamber divided by longitudinal fins	(1) Continuous (2) Reciprocating iron conveyor	(1) Continuous (2) Screw conveyor in retort	(1) Continuous (2) Through each retort in succession by screw conveyors	(1) Continuous (2) Gravity and screw conveyor at bottom of retort
8 Other mechanical appliances	Reciprocating scraper to prevent clogging		Material charged into each container by piston. Retort revolves by internal C.I. roller gearing with fins		Revolving scraper fitted if required		Vapour outlet valve opens when pressure exceeds 5 lb. per sq. in., and is closed under smaller pressure
9 Method of cooling carbonised charge	Quenched by hand with hose-pipe		Cooling chamber	Discharged through water seal	Cooling zone in retort	Cooling chamber	Cooling chamber
10 Heating arrangements : Fuel employed	Gas	Gas	Gas	Gas	Gas	Gas	Superheated steam
11 Method of burning fuel	Burners	Burners	Burners	Burners	Burners	Burners	
12 Application of heat to charge	External	External	External	External	External. Gas passes through hollow screw conveyor	External	Internal
13 Control of heating zones				Yes			
14 Utilisation of heat of exhaust heating gases				Regenerative setting			
15 Use of steam	No		No		No	Yes	Yes
16 Location of outlet for gaseous products	Lower end of retort	Discharge end of retort	Discharge end of retort	Channel in roof of retort leading to hydraulic main at feed end	Several outlets along top of retort	Outlet from bottom header	Top of retort
17 Gas pressure in retort	Slight vacuum	Slight vacuum	Slight vacuum		Slight vacuum	Slight vacuum	Max. 5 lb. per sq. in. Varying pressure.

between several different branches of science and technology, but other branches, such as geology, botany, etc., are likely to afford valuable information in more limited directions.

Some of the many patents which have been taken out have, however, passed from the inventive to the experimental stage, while a few have gone a step further, and have been built on a scale in which, if successful, they might be reduplicated as units of a possible commercial plant. By means of others, again, it has been possible to place on the market small supplies of coke and oil, but time alone can show whether the commercial results obtained are such as to ensure their survival. The authors have studied in detail some forty of the most promising of the British, American and German retorts, and the following broad generalisations of their most distinctive features are given.

CLASSIFICATION BY METHOD OF HEATING

In the broadest sense it has already been pointed out that there are two methods of heating by which the problem of low temperature carbonisation may be attacked; but the fact is not overlooked that a process may be successful in which these two methods of heating are combined :—

(1) The coal may be distilled in a gastight retort in which the whole of the heat for carbonisation is passed through the iron or fireclay retort wall. The fuel, solid, liquid or gaseous, is burnt either in a large chamber which almost completely surrounds the retort itself, or in a succession of smaller heating chambers built on the outside of the retort wall, i.e. external heating.

(2) The coal may be exposed to the action of heat derived from the sensible heat of some substance which is mixed with the material to be carbonised. The substance forming the heating material is usually a large amount of producer gas, either generated within the retort itself, as in the Maclaurin type, or externally, as in the Nielsen retort. Proposals have, however, been made from time to time to use materials other than producer gas for this purpose, these proposals including the use of superheated steam, heated low temperature coal gas, melted lead or hot sand, i.e. internal heating.

Since one of the main difficulties associated with the problem lies in conducting heat into the interior of the mass, classification under the headings mentioned above seems to be the most important. In the first case, the coal must be carbonised in thin layers. This renders difficult the design of a plant which shall have a large capacity, and, at the same time, be compact and occupy a minimum of ground space. It is simple, however, to obtain and recover the maximum of products,

especially the gas, both as regards quantity and quality. In the second case, the introduction of heat is much facilitated, but when producer gas is used as the heating vehicle, it involves dealing with 20,000 to 25,000 cubic feet of a low-grade gas, having a calorific value of about 230 B.Th.U. per cubic foot as a maximum. If badly designed or carelessly worked, the heating value of the gas may be greatly reduced below this figure.

If superheated steam is used, either alone or in conjunction with producer gas, as the heating medium, the quality of the gas available for sale may be greatly improved, since the steam will be condensed out in the cooling plant, whereas producer gas passes along as a diluent. Unless steam, however, can be obtained more or less as a waste product from some other operation, the difficulty of recovering and making use of its high latent heat renders it expensive as a material whose only function is to distribute heat which it possesses by virtue of its temperature only.

Although most types of retort may be classified according to the considerations quoted above, we find examples which embody features of both classifications. For instance, when a small amount of steam is used at the low temperatures here considered, it has been previously seen that decomposition of the steam by carbon is almost negligible, and the action of the steam is probably mechanical, not only in sweeping away the vapours of carbonisation, but at the same time in assisting the distribution of heat by convection. The fact, therefore, that an externally heated retort is steamed brings it partially within that group associated with internal heating.

CLASSIFICATION BY METHOD OF WORKING

Having arrived at a basis of classification on the broadest lines, it is obvious that many subdivisions may be made within these two groups. One of the most important of these is whether the process is continuous or discontinuous in action. The advantages of a continuous method of working are :—

- (a) Conservation of labour and heat.
- (b) A tendency to increased life in the retort, owing to constant temperature conditions being maintained in the various parts of the apparatus.

    On the other hand, it may be difficult to recommence operations in the event of an interruption or breakdown.

The advantages of an intermittent process are :—

- (a) Mechanical arrangements may sometimes be simplified.
- (b) Examination of the interior of the apparatus is simplified.
- (c) The charge is under more direct control.

In comparing these two systems it is significant to note that only about 10 per cent of the processes examined are intermittent in their working. In gasworks, however, the intermittent horizontal retort is extremely popular.

OTHER METHODS OF CLASSIFICATION

Other methods of classification which naturally suggest themselves are, according to the position of the retort axes, vertical, horizontal and inclined, according to whether the retort moves or not, according to the material of the retort, and many others. These, however, while useful for descriptive purposes, do not call for any special comment.

CHAPTER XVIII

A DISCUSSION OF SOME DIFFICULTIES ENCOUNTERED IN LOW TEMPERATURE CARBONISATION

A. RETORT MATERIALS

MATERIALS USED IN THE GAS-MAKING INDUSTRY

In all processes involving the application of heat, it is necessary to devote careful attention to the material through which the heat is passed from the heating agent to the material under treatment. In the earliest gas-making processes it was customary to use retorts made of cast iron. Murdoch (in 1792, at Redruth), who was the founder of the gas-making industry, first manufactured gas in a cylindrical iron pot, closed by an iron lid, and set over an open fire. Difficulty was encountered in discharging this retort, and at the beginning of the 19th century Murdoch adopted for his larger experimental work a horizontal iron retort of cylindrical form, which solved some of the difficulties in this direction, since the charge could be raked out after carbonisation. These retorts were small in size, the maximum at this period measuring about 20 inches diameter by 7 feet in length.

Murdoch, whose first experiments were carried out at very low temperatures, made an early discovery that it was necessary to adopt higher temperatures if an increased gas yield was to be obtained. The difficulties attendant upon the carbonisation of large masses of coal were also soon apparent, and attempts had to be made to overcome these disadvantages. At length the fact was recognised that, at the low temperatures employed at that time, it was essential to carbonise the coal in thin layers. Although this principle was well recognised in the early days of the gas industry, it is interesting to note that many of the inventors, who have since 1906 turned their attention to low temperature carbonisation, have failed to utilise the experience gained as early as the first half of the last century.

Following the progress of the gas-making industry, we find that by the middle of the 19th century, the practice of manufacture was confined to the use of retorts of oval or �⎕-shaped section.

The development of the ordinary process of gas-making has, since its inception, followed the line of obtaining the greatest possible quantity of gas of suitable illuminating or calorific value, produced by means of increase of temperature. In the earlier retorts it was impossible to use temperatures in excess of $800°$ C., and even then the deterioration of the material proved so rapid that the life of the retort extended only over a few months. Growth of cast iron, even at lower temperatures than those quoted above, was very serious. It is only of late years that scientific research has been the means of explaining this phenomenon, which has been for many years a familiar difficulty in certain industries. At temperatures exceeding $650°$ C., unless great care is taken in the selection of the metal, it is

239

possible for a cast iron twelve-foot retort to increase several inches in length within a comparatively short time.

The troubles attending the use of metal retorts at the higher temperatures led the gas industry to experiment upon retorts made of fireclay, and these came into common use about 1850, their section being much the same as those of the earlier cast iron retorts. After the inevitable troubles associated with such a radical change in material had been overcome it was possible for the gas industry to increase the temperatures to a much greater degree. These temperatures at the present time average from 1200° C. to 1300° C., and it is interesting to note in passing that the successful use of refractory materials at these temperatures has only been rendered possible by the application of scientific knowledge regarding such factors as their melting and softening points, thermal expansion, both permanent and temporary, and other details.

It has sometimes been suggested by those who are not fully conversant with the subject, that all that is necessary in securing effectively the low temperature carbonisation of coal is to return to the practice of the early part of last century. A very brief consideration of the question, however, proves the fallacy of this proposal. It is true that the temperatures utilised in common practice at that time were only to a small degree in excess of those likely to give the most favourable results in a process which aims not at gas production, but at the production of the greatest quantity of oil, in addition to a coke containing some 8 to 10 per cent of volatile matter. At the time when metal retorts were in general use in the gasmaking industry, coal and labour were alike cheap, and the prices obtainable for gas were relatively in excess of those commercially possible to-day. Although it is possible to utilise the experiences which have been gained by the gas industry, it would be absolutely impracticable under modern conditions to establish an industry capable of carbonising the tens of millions of tons of coal at present used in the raw state by merely setting back the clock of progress in the gas industry by half a century or more. On the contrary, success in this national problem can only be brought about as the result of careful study of the whole matter, by reasoned judgment, and by experimental work based on the partial successes and failures of the past.

ADVANTAGES OF METAL RETORTS

In the design of any form of retort or oven, one of the first questions to be determined is whether to adopt metal or refractory material for the actual walls through which the heat has to make its way. Both materials present certain advantages and disadvantages; and, in the case of carbonisation at about 600° C., neither possesses properties which would rule it out, as would be the case were metal proposed for high temperature work.

One advantage of a metal retort lies in its high coefficient of conductivity for heat. This advantage is accentuated by the fact that the walls may be kept down to a minimum thickness. Another point of superiority which the metal retort possesses over that made of refractory material is the ease with which it may be made and kept gastight. Leaks are not likely to develop, and, even should some failure of the joints ultimately occur, the escape of gas, either in the inward or outward direction, will probably be small. The leaks can also be repaired with comparative ease during a periodical overhaul.

CAST IRON RETORTS

In the case of cast iron, one of the difficulties which may presen possibility of growth at the temperatures used, especially in the presence of super-heated steam. The distortions to the setting, which are serious if not properly provided for, can be reduced to a minimum by allowing means for the changing levels of points in the retort to adapt themselves to the levels of the surround-ing brickwork. The present-day knowledge of the properties of cast iron is also much greater than that of the middle of the last century, and mixtures can now be provided in which growth is reduced to a minimum. In a paper by Rugan and Carpenter,[1] before the Iron and Steel Institute in 1909, the problem is taken up in a comprehensive manner. Engineers and others interested in the use of metal which is required to withstand high temperatures, will find this paper a veritable mine of information. In a further paper by Prof. Carpenter,[2] read before the Staffordshire Iron and Steel Institute, a summary is given of the state of know-ledge practically as it exists at the present time, and the authors cannot do better than quote Prof. Carpenter's own words as follows :—

" The chief sinner in the swelling of grey irons on heating, whether at high or low temperatures, in air or furnace gases or superheated steam, is unques-tionably silicon, present not as such, but as dissolved iron silicide in the main bulk of the iron. But this silicide can exert no influence whatever in producing growth by separation as silica and iron oxide, unless graphite is present, which, on heating the iron, allows oxidising gases to penetrate into and gradually attack the metal chemically. Without graphite silicon is absolutely innocuous"

From his experiments Prof. Carpenter draws certain conclusions which may be summarised as follows :—

(1) Any grey cast iron, if heated either repeatedly or for prolonged periods,

[1] Rugan and Carpenter. "The 'Growth' of Cast Iron after repeated Heatings." *Journ. of Iron and Steel Inst.*, 1909, **2**, 29.
[2] Carpenter, Prof. H. C. H. "Further Notes on Cast Iron Growth." *Staffordshire Iron and Steel Institute*, Dec. 15, 1916.

Q

either in superheated steam above 250° C. or in air or furnace gases above 650° C., will swell and eventually crack.

(2) The amount of growth will depend on the chemical composition and constitution of the iron and may be expected to be least for low silicon cold blast Staffordshire iron, and most for high silicon hot blast irons.

The solution of the growth difficulty put forward by Carpenter is that no iron containing free carbon should be used ; and, further, that even when an iron contains no free carbon, it should not deposit it on repeated or prolonged heating.

Campion and Donaldson[1] record experiments upon the changes of volume,
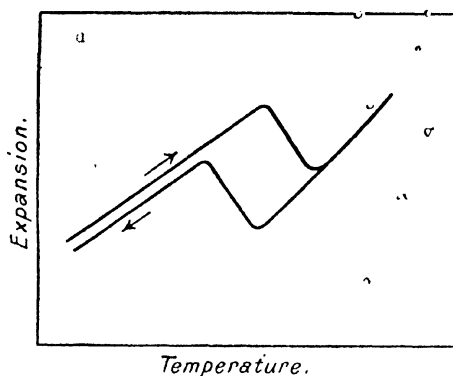
FIG. 50.—THEORETICAL EXPANSION CURVE OF STEEL
FOR THE RANGE 600–900° C.

weight and strength of cast iron after repeated heatings to 550° C. They found that the experiments at 450° C. on the changes of volume due to alternate heatings and coolings gave erratic results with no tendency to change in any particular direction. Heating up to 550° C., however, caused a growth which continued after fifty-five heatings. The strength of the iron was also seriously reduced by this treatment. Experiments on the tensile strength of cast iron at various temperatures revealed a well-defined maximum strength at a temperature of about 400° C., the strength falling very rapidly at higher temperatures.

The growth of cast iron at high temperatures may be illustrated in another way. It is well known that 600–1000° C. is a critical range for the expansion of iron and steel. Between these temperatures the steel contracts during heating over a certain range and expands during cooling over another range. The theoretical expansion curve of steel for a range 600–900° C. during heating and cooling is

[1] Campion and Donaldson. "Some Influences of Low Temperature on the Strength and other Properties of Cast Iron." Trans. Inst. of British Foundrymen, June, 1922.

illustrated in Fig. 50. The expansion curve for cast iron is, however, very different. Experiments made by Souder and Hidnert[1] on a specimen of cast iron which contained 3·08 per cent carbon and 1·68 per cent silicon gave an expansion curve which is shown in Fig. 51. The difference between the heating and cooling portions of the curve is in this case almost entirely due to the growth of cast iron at high temperatures. It is seen that the growth becomes marked at temperatures of about 600° C. and upwards. Souder and Hidnert give the coefficient of expansion
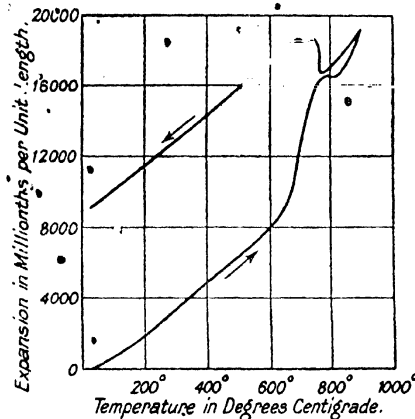
Fig. 51.—EXPANSION AND GROWTH CURVE FOR CAST IRON
(SOUDER AND HIDNERT).

for cast iron between 25° and 300° C. as 11·6 × 10⁻⁶, but state that the values found at high temperatures must be considered as approximate only, as doubtless they contain a percentage of growth in addition to expansion.

In his paper to the Iron and Steel Institute in May, 1911, Prof. Carpenter[2] gives the following approximate composition of a cast iron in which the growth is likely to be a minimum :—

		Per cent.
Carbon	. . .	2·25-2·5 (no free carbon)
Silicon	. . .	0·5-0·6
Manganese	. about	1·5

A further valuable property of this material is that repeated heating substantially improves its tenacity and probably its ductility also. It should, however, be

[1] Souder and Hidnert. " Thermal Expansion of a few Steels," Paper No. 433, U.S.A. Bureau of Standards.
[2] Carpenter, H. C. H. " The Growth of Cast Iron." Journ. of Iron and Steel Inst., May, 1911.

initially heated very carefully on account of its liability to crack during the early heats.

In deciding whether or not to adopt cast iron for the retort material in a low temperature process, it will be seen that, so long as 650° C. is not exceeded, trouble from growth is not likely to be serious if care be taken to ensure compliance along the broad lines which have been determined by Carpenter. The main difficulty in doing this, however, at the present time is that of ensuring that any specified mixture is actually used, coupled with the reluctance shown, by ironfounders in accepting orders to specifications such as these, which differ from their usual practice. This difficulty, however, while a very real one in the execution of experimental work on carbonisation to-day, will undoubtedly disappear when such retorts are required in large quantities, and attractive orders can be offered to the makers of castings. In the case of some large cast iron retorts recently erected at H.M. Fuel Research Station for carbonisation at 600° C., a suitable material was obtained by placing the order with a foundry firm who were accustomed to supply similar retorts for the Scottish shale industry.

The advantage which cast iron presents over mild steel for retorts is its greater rigidity at the working temperatures and its greater resistance to influences of a corrosive nature. The fact that it can be cast and therefore reproduced in quantity with a minimum of labour is also of importance.

MILD STEEL RETORTS

In some cases retorts have been made from mild steel. This material was not available to the pioneers of the gas industry. so that no data are at hand gained from their experience. Provided the temperature is not too high, mild steel offers many advantages. It does not possess the property of growth, but on the contrary contracts slightly after repeated heatings. Moreover, retorts of varying shape can be put together with ease although the labour cost of manufacture is usually higher than that of cast iron, when once the pattern has been constructed, owing to the many joints which must be made. In a slightly oxidising or reducing atmosphere mild steel offers considerable resistance to corrosion. An interesting example of this is given in the accompanying photograph (Fig. 52) of one of the horizontal retorts at H.M. Fuel Research Station. This retort was removed from the setting after having been under heat more or less continuously for eighteen months. No trace of scaling or corrosion can be detected, and the retort appears to be in exactly the same condition as when it was first installed. Some of the remaining retorts have, at the time of writing, been in almost daily use for some three years, and although a close examination, such as could be made in the case of the retort illustrated, is impossible, they do not appear to have suffered in the slightest degree.
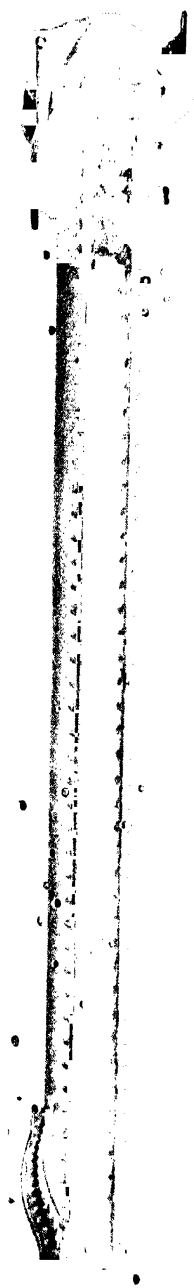
Fig. 52.—Photograph of Mild Steel Retort after Service.

One of the most serious disadvantages attending the use of mild steel is its liability to soften if a working temperature of 600° C. is exceeded. This softening occurs somewhat suddenly, and necessitates the careful watching of the heat; but since efficient temperature control is necessary in any modern carbonising plant, such a disadvantage can readily be overcome. Mild steel shares many of the advantages attending the use of cast iron, since retorts of this material can be made tight with ease in the first instance, and can be maintained in that condition without much difficulty.

The foregoing remarks on the suitability of mild steel for low temperature retorts only apply when the material is not subjected to heavy stresses. At the temperatures usually employed the elastic limit of the material is low, and, moreover, the metal is liable to a continuous creep with time under load at stresses much below those usually quoted for the yield point at any given temperature when tested in the usual manner in a testing machine. Difficulties, therefore, may arise in cases where it is desirable to maintain accurate registration in the working parts under full heat, especially when it is necessary to use comparatively high stresses (say above two tons per square inch). This fact accounts for many of the difficulties which have in the past been experienced in the classes of apparatus where the coal is mechanically propelled through the retort by a chain conveyor or other similar device, and has often led to the abandonment of the method.

ALLOY STEEL AND ALLOY CAST IRON RETORTS

Some of the difficulties attending the use of steel for carbonising machines could be overcome by the adoption of steels alloyed with elements such as nickel, chromium or tungsten. Alloy steels of this description have been extensively utilised for the valves of motor and aeroplane engines and for many other applications where the conditions are even worse than those which are likely to obtain in low temperature carbonising machines. These metals are, however, expensive, both in their production and in the difficulty which is frequently experienced in their machining. In common with many other engineering problems, the commercial solution of low temperature carbonisation would be greatly facilitated were it possible to produce at a price little greater than that of mild steel, a metal which would maintain a good degree of rigidity at temperatures in the neighbourhood of 750° C.

In this connexion Professor Carpenter has suggested to the authors the possibility of using an alloy cast iron, as, for example, a cast iron containing a small percentage of chromium. There is some reason to think that, amongst such alloys, one or more may be found which are equally satisfactory from the standpoint of growth, but are less brittle than the cast iron whose composition is given on page 243,

and which have an increased strength at high temperatures. From a manufacturing standpoint, a further advantage, of a chromium cast iron at least, would be that such an alloy could be produced from a chromiferous iron, possibly without the necessity of adding an alloying element.

The above suggestion is, however, tentative only ; but there are good grounds for belief that research in this direction is likely to lead to useful industrial results.

FIRECLAY AND REFRACTORY MATERIALS

As has already been pointed out, the use of refractory materials for retorts has long been universal in gasworks. The conditions are, however, very different from those of low temperature carbonisation, since the temperatures used in the former are almost double those of the latter. The use of refractories, however, presents some attraction to the designer of low temperature retorts. He is working his apparatus at temperatures which cannot have much effect upon the settings, his margins for safety are higher and, although in general his retorts are likely to be more expensive than similar retorts of cast iron, there is no necessity for his embarking upon the use of the high-class and expensive materials which are essential when temperatures of 1200–1300° C. are used. Against this must be set the comparative lack of conductivity of the material, necessitating the use of higher flue temperatures. Another disadvantage lies in the comparative ease with which a fireclay retort may be damaged by heavy or careless poking. The fireclay retort also is a more delicate structure than one of cast iron, and one which is essentially much more prone to develop leaks. The question of leakage is one of extreme importance in low temperature carbonisation. In high temperature work the yields of gas per ton are at least four times those obtainable in the subject under consideration. Again, low temperature gas, although small in volume per ton, is very high in calorific value. It follows, therefore, that a small amount of leakage is proportionately more serious and less under control in low temperature work than in high temperature work. Another grave difficulty attending the use of leaking retorts lies in the maintenance of uniform heating. If a slight vacuum be put on the retort, there will be danger of the leakage of flue gas into the retort from the flues increasing the inert gases and increasing the volume to be dealt with by the condensers and scrubbers, these factors bulking seriously owing to the small volume per ton of the gas made. If a balanced pressure or a positive pressure be used, there is great danger of rich gas escaping into the flues, and being burnt there. This may be the cause of serious local overheating, but perhaps a more undesirable feature is that this condition causes the operator to lose control of the temperatures. One of the authors has actually seen retorts at work when the leakage outwards has been so serious as to necessitate the almost complete shutting

off of the heating gas from certain of the combustion chambers. It is obvious that such a state of things may become a grave source of danger in the operation of the plant, and that this question of leakage must be seriously faced by the designer if success in low temperature carbonisation is to 'be attained.

Before leaving this question of leakage it may be well to consider briefly one argument always brought forward by advocates of refractory retorts for low temperature carbonisation, viz. the tendency of leakage to remedy itself owing to deposit of carbonaceous materials in the cracks. While this is undoubtedly the case in high temperature practice, it only occurs in a very minor degree—if at all— in retorts worked on a low temperature system. In the Scottish shale industry it is a well recognised fact that leakage does not tend to cure itself in the manner described. This is probably due to the different nature of the oils obtained.

CHOICE OF METAL OR FIRECLAY RETORTS

The above considerations present some of the more important factors which have to be faced at the outset in the design of a low temperature plant. While it would be inadvisable to form too strong an opinion as to the relative merits of metal or fireclay retorts without much more experience than is available at the present time, it appears to the authors that a considerable balance of advantage lies with the metal retort, and that those manufactured of a suitable mixture of non-growing cast iron are likely to prove the most successful and also the most reliable. Retorts of steel are also good, but, as has been emphasised above, there is a risk of damage by overheating, with consequent distortion and sagging, should the temperatures get out of control from carelessness or any other cause. The authors entirely fail to see that any advantage has been obtained by the adoption in certain cases of refractories as the material for retorts, while unfortunately the disadvantages have frequently been only too obvious.

The above considerations are mainly based on the type of apparatus heated from the outside, when the whole of the heat necessary for carbonisation has to pass through the walls. Where internal heating is used, the conditions may be different. We are no longer faced with the difficulty of passing heat through a great thickness of refractory material, and it may quite well happen that a form of retort built of brickwork with a fireclay lining will prove the most suitable. In this direction, however, generalisation is impossible ; a contrast might be made, for example, of two processes, both involving the same principle, viz. carbonisation by hot gases. In one of these—the Maclaurin retort—the heat is generated by partial combustion in the lower layers of fuel in a retort of the producer type, and carbonisation is effected by the hot gases from the lower zones passing through a great thickness of fuel bed, with the result that the upper layers of fuel are distilled at a low

temperature. For this process Maclaurin adopts a retort or producer, built of brickwork, and lined with refractory material, this probably forming the best method in this particular case. In the Nielsen retort, on the other hand, in which exactly the same principle is used, the hot gas is obtained from a separate producer. This hot gas is passed through a long horizontal rotating cylinder, through which the coal travels in the reverse direction. In the latter case, it is obvious that a complete firebrick arrangement would be impossible, and that it is essential for the shell of the horizontal cylinder to be composed of metal

B. HEAT TRANSFER FROM HEATING MEDIUM TO MATERIAL UNDER TREATMENT

FLOW OF HEAT THROUGH RETORT WALLS

In all types of retort used for carbonisation purposes, it has long been a matter of considerable doubt as to what is the temperature at various points in the charge when a constant and known temperature is maintained in the gases present in the heating chambers, and also what amount of heat can be transferred from the gases which form the heating medium to the material under treatment, when both are at known temperatures. The problem is one of extreme complexity, and presents itself in many other directions, such as heat loss from the walls of buildings and the transfer of heat through boiler plates.

In supplying heat through a retort wall it is first necessary to transfer the heat from the gases in the heating or combustion flues to the outer surface of the retort wall, then to pass the heat through the retort material to the inner surface and from thence to transfer the heat to the coal which may be lying more or less in contact with it, or at some distance away, in which case the heat has to be again transferred to the coal by conduction, radiation and convection through the gases present in the retort.

The ordinary methods for the determination of the coefficients of conductivity for different materials depend on maintaining the two surfaces of a test piece under constant and known temperature conditions. Such coefficients are not intended to take into account the temperature differences between the heating and cooling agents and the surfaces with which they are in contact which are necessary, to ensure a given heat transfer. In the absence of much additional information, therefore, such coefficients do not afford any exact data as to the amount of heat per square foot of heating surface which will flow from the heating source to the material under treatment. Broad generalisations may be made, however, such as the fact that the temperature difference necessary will be very much less when the wall is thin and has a high coefficient of conductivity than will be the case

when the wall is thick and is made from a material of low conductivity. In certain isolated cases, however, useful working data have been deduced from works operations for the overall coefficient of heat transfer through firebrick walls. Sir George Beilby, from experiments at the lower temperatures, determined the figure of 11·7 B.Th.U. per hour for each degree Centigrade of temperature difference between heating and cooling materials at the two sides of the brick, the temperature difference being of the order of 200–300° C. Lewes,[1] on the other hand, was of opinion that, " at the ordinary working temperature of a retort under gasworks conditions, the amount of heat transmitted approximates to 25 B.Th.U. per square foot of surface for each 1° C. difference in the temperature of the outer and inner surface of the retort, and that this is not seriously affected by the thickness of the fireclay, as conduction is so slow with a retort three inches thick that it is probably only the internal portion that is cooled to any great degree when a fresh charge is fed into a properly heated retort, and the mass of fireclay acts as a store of heat, so that the heat has only a short travel."

The differences in such typical figures as are quoted above are easily accounted for by variations in such factors as surface conditions, including degree of exposure to convective effects of gas currents. The amount of contained air is also well known to affect the apparent conductivity of any material. The degree to which this air is free to travel about within the substance, or, in other words, its permeability, must also affect the experimental figures to a marked degree. There are also wide variations due to the material itself. In general, fireclay bricks possess a higher coefficient of apparent conductivity than does silica. Again, it is affected by the temperature of burning, the higher temperature contributing to higher coefficients. Lewes quotes for an ordinary fireclay burnt at 1050° C. a coefficient of conductivity of 1·07, which increased to 1·81 with a temperature of 1300° C. He also points out that this explains why an old retort will give better carbonising results than a new one, provided it is sound.

Other general results of investigations on the thermal conductivity of refractory materials may be thus summarised :—

(1) The thermal conductivity of silica and fireclay materials increases with the temperature, but that of magnesite bricks decreases.

(2) The thermal conductivity of silica and fireclay materials, although varying with different compositions, is very similar, that of fireclay being the greater. The thermal conductivity of magnesite is approximately twice that of fireclay, and that of carborundum at least five times that of fireclay.

When metal is used for the retort wall, some of the uncertainties associated with the transfer of heat disappear. The thermal conductivity of a metal is many

[1] Lewes, Vivian. *The Carbonisation of Coal.* (Benn Bro'

times greater than that of any refractory material, and it is also possible to work with a much smaller thickness. These two factors working together make it inconceivable that the interior of the retort wall should be more than a few degrees less in temperature than the outside, and the heat transfer is therefore governed almost entirely by the rapidity with which the heat can be supplied to the outer surface by the flue gases, and abstracted from the inner surface of the metal by the charge undergoing carbonisation. The thermal conductivity of the metals is well known for temperatures not greatly in excess of atmospheric.

The variation in the thermal conductivity of iron at the higher temperatures is, however, somewhat uncertain. Most of the observations taken by experimenters do not cover wide temperature ranges. Jaeger and Liesselhorst[1] observed a slight fall in the conductivity of pure iron from 0·161 to 0·151 as the temperature rose from 18° C. to 100° C. At temperatures between 54° C. and 102° C. Callendar[2] found a slight fall in the conductivity of cast iron from 0·11¼ to 0·1¼2. Experiments on the thermal conductivity of cast iron are described in the *Physical Review*, March, 1922. It is stated that, between 197° C. and 545° C., the thermal conductivity rose from 0·0718 to 0·2050. It must be remarked, however, that the value at 197° C. appears to be very low, and the variation over the range of temperature large.

Such then are some of the difficulties which beset the designer who wishes to obtain figures for heat transfer in the complex process under review. These difficulties are intensified when the heat transmission through the mass of coal itself is considered. The transmission may, and is indeed likely to vary widely, according to the size of coal, the grading of size of coal, the behaviour of coal on carbonisation, whether the conditions and character of the coal tend to the formation of sticky masses, foaming, etc. ; and, although a certain amount is known about the heat transmission at high temperatures, much further experience and scientific investigation is yet required of the phenomena of overall heat transmission at the temperatures usually proposed for low temperature carbonisation.

C. CHARGING AND DISCHARGING

Of the many difficulties which beset the earlier workers in low temperature carbonisation and which in many cases are by no means yet overcome, the easy charging and discharging of the retort is one of the most important. In the case of the coals of low coking power the material may be passed with ease through many forms of retort. In this case, however, another difficulty usually presents itself, in that the coke is generally in the form of breeze, owing to the lack of proper cohesion, which, while making for easy retorting, does not endow the coke with sufficient strength to withstand the pressures and blows associated with its move-

[1] Jaeger and Diesselhorst. *Wissenschaftl. Abhandl, physt tech. Reichsanal*, 1900, **3**, 26.
[2] Callendar. *Phil. Mag.*, 1899, (v), **48**, 519.

ments both in the retort and in the discharging operation. Even in cases where
the pieces of coke are of the same form and size as the corresponding pieces of
coal charged, the coke does not usually possess sufficient strength to form a
marketable produc without further treatment, such as briquetting, with a small
amount of pitch. This fact is well established by the experience of Sir George Beilby
with the Maryhill retort, already described on page 198.

It will be seen also from the parallel experience of the Scottish shale oil industry,
that there is no particular difficulty in passing a non-caking material through a
retort of simple character ; and it is significant to note that inquiry into the results
presented by some enthusiastic inventors when first bringing their appliances
before the public, shew that they frequently base their claims upon tests executed
on shales or non-caking coals. Little is said as to the results when a wider
variety of coals is utilised.

The difficulties in working associated with caking coals usually resolve them-
selves into two forms : firstly, the trouble caused by the passing of the coal through
a fusible stage, with the resultant agglomeration of the material into pasty masses,
tending to adhere to the surrounding retort walls, and secondly, the effects due to
the so-called expansion of the coal on carbonisation. It has already been pointed
out that this expansion or increase in bulk of the carbonised mass is due to the
size of the bubbles which are blown in the fused material by the gas given off by
its decomposition. The effect of treatment on the formation of different types
of bubble structure is a matter about which further experimental work is desirable.
The subject is of fundamental importance in many branches of the carbonisation
industries, affecting makers of blast-furnace coke and others with whom we are
not concerned in the present volume.' It is, however, evident that the fusibility
of a coal may affect a retorting plant in two ways. The first difficulty is that the
working parts become clogged and gummed up, after which, when coking is com-
plete, the whole apparatus is reduced to much the same state as a piece of ferro
concrete, the coke representing the concrete, and the worms or other metal working
parts of the retort, the iron reinforcement. The advocates of methods utilising
worms apparently held the opinion at first that all that was necessary in order to
feed a pasty mass through a tube was sufficient power together with a sufficient
strength in the working parts. Unfortunately, owing to the subsequent coking
of the plastic mass, this did not work in practice, and plants with such devices
frequently stopped completely after working for a few hours.

The other difficulty associated with attempts to carbonise a caking coal is
exemplified by the earlier experiences with the Parker process. Here coal was
carbonised in a retort bench consisting of vertical iron tubes heated from the exterior.
The " expansion " of the carbonised mass caused the material to wedge itself firmly
against the walls of the tubes and much labour had to be exerted before such

retorts could be discharged. Replacing the tubes with others having a greater taper only results in slightly easing the process of discharge, but does not solve the difficulty. A number of expensive failures in the past may be traced to this cause. Indeed, cases are known where actual rupture of the retort walls and the consequent wrecking of the plant have occurred.

It will be well at this stage to pass in review some of the methods which have been suggested from time to time for overcoming the two types of difficulty, which are closely interrelated, although for purposes of convenience each are considered separately.

In the type of retort in which a screw conveyor is used to propel the carbonising mass through a metal tube, acting as the retort proper, the difficulty first mentioned appears. As long as non-caking coals are used, the material progresses as the screw is rotated. When caking coals are used, however, there is a zone of temperature during which the material gets into a more or less pasty condition, and tends to stick to the screw thread. If this should happen, the material rotates with the thread, and does not progress along the retort. in consequence the material between that point and the point of entry is held up. Furthermore, the resulting increase in the frictional resistances requires an augmented power to keep the conveyor rotating.

Various scraping devices have been suggested to obviate this difficulty, of which a double screw is perhaps the most typical. In this case, the screws have the same hand and equal pitches, but rotate at the same speed. On a closer examination, however, it is found that two screws under these conditions do not have a true scraping action. Since each thread must contain an amount of material which will ensure an appreciable throughput for the retort, there must be a relatively large difference between the diameters of the screw at the crest and root of the thread. That is to say, the thread angle at the crest is considerably less than the thread angle at the root. Although it would appear, from a cross-section drawn with the two threads in this position, that intimate contact occurs throughout the thread faces, the screws could not be placed in this position on account of the interference of one thread with another in the planes adjoining the axial plane. A double thread is therefore no solution to this particular problem, since the pasty mass would eventually stick to both screws, and the throughput still be held up.

It should be noticed in passing that, in the Marshall–Easton process described on page 227, in which a double conveyor of this type is utilised, a claim for a scraping action is not made by the inventors. In this case the screws are utilised to control the speed of the material through the retort, and the claim is made that highly bituminous coal in the sticky stage will be wiped forwards and downwards by the relative motion of the screws.

Before leaving the subject of foaming and expansion of fusible coals, it will be of interest to draw attention to a method for overcoming this suggested by Mr. Harold Hird,[1] in which an attempt is made to reduce the size of the bubble structure by inserting a suitably shaped bar down the centre of the charge in an intermittently worked vertical retort. This bar is kept in slow rotation during the carbonisation period. The idea underlying Mr. Hird's invention is one which is well recognised, viz. the desirability of providing a free outlet for the gases as soon as they are formed, and at the same time of preventing their coming in contact with highly heated surfaces, or of their passing through zones of temperature higher than those at which they were formed. As the rod rotates in the centre of the coal, it ensures that a loose core is provided, through which the gases can pass with ease. As carbonisation proceeds, this loose core is gradually developed into a hole by the pressing down of the loose material into the surrounding coal substance. Since carbonisation proceeds from the outside towards the centre, the sticky annulus, in which the bubbles develop, moves gradually inwards in any form of externally heated intermittent apparatus. If the gases developed in the sticky annulus are not removed rapidly the structure will tend to be loose and open; that is to say, unless external pressure is placed on the coal, or an inert material provided, such as a non-coking coal or coke, in order to absorb the excess of the binding material. In the method mentioned it is possible that a much easier path for the escape of the gas from the fused zone will be provided than is the case with many other devices which have from time to time been suggested or tried with the same object.

A method of providing a mechanical feed through a horizontal tubular retort which appears to be promising is that of breaking the screw thread up into a series of paddles, as is done in the first stage of the Carbocoal process, described on page 210. Here agglomeration into unmanageable blocks is prevented by the motion of the paddles, while a continuous movement of the material in the forward direction is induced by the angle to which the blade is set. In the Burney retort a solid screw is provided, in order to urge the material in the forward direction, while a series of scrapers, which taken together form what might be termed the skeleton of a screw, are arranged to keep the main screw clean and prevent undue agglomeration.

Other modifications of this principle are exemplified in the Freeman retort. Here we have a succession of heated annular shelves, set one above the other in a cylindrical vertical column. To any one shelf a feed of coal is provided either at the outside or inside of the annulus, according to its position in the series. The material undergoing carbonisation is caused to travel in a circular direction by a series of rotating scrapers. These scrapers are set at a slight angle, so that a radial

[1] Hird, Harold F. Pat. Spec. No. 184,525 (1922).

component is given to the approximately circular path. In this way the coal is gradually fed from one circumference of the annular shelf to the other and then falls to the next shelf in the series.

In the Fusion retort we find a horizontal tube which rotates. The material to be carbonised is fed into one end of the tube and extracted at the other. The retort is horizontal, and the feeding principle depends purely upon the tumbling action of the coal due to the rotation. When fusible coals are used, an arrangement of star or cruciform bars is inserted. These bars, as the retorts revolve, will turn over, and it is claimed that the chipping action caused by the bars as they fall into contact with the retort shell, is quite sufficient to prevent the coal from permanently adhering to the wall.

The retorts discussed above are all of the type in which heat is passed into the material through the wall. In the internally heated "Sensible Heat" (Nielsen) retort, described on page 231, it will be seen that it is possible to use a coking coal, and that this coal passes along without any apparent difficulty, although agglomeration of the carbonised mass undoubtedly takes place, since a large proportion of the smokeless fuel, produced from a highly fusible coal, is of a size much greater than the slack from which it was made, besides being dense and fairly hard. The product also presents the appearance of having been balled up and rolled, in much the same way as a baker rolls a loaf of bread. The inventor attributes the freedom of the retort from agglomeration and accumulation on the walls to the dry dust which is passed in along with the hot producer gas used for carbonisation. It is, of course, possible that this dust acts in the same way as the dusting of flour, which prevents the dough from adhering to the pasteboard in the analogy quoted above.

Let us now turn to the other aspect of the question, viz. that concerning the so-called expansion of the finished product. This difficulty manifests itself in the most serious form in those types of retort in which enclosed, or partially enclosed, chambers are completely filled with the coal to be carbonised. It must be remembered that some pressure during the plastic stage is of advantage, since it tends to the reduction in size of the bubble structure, and if provision is made for the easy release of the product, it is likely to be denser and harder than if it were given complete freedom to expand during the critical period. In the Barnsley retorts, described on page 206, this end is accomplished by the provision of the collapsible plates which run down the centre of the retort. In the Pringle and Richards retort, which is of the type where a series of moulds is arranged along a conveyor, the latter passes through the heated zone of the retort. With fusible coals, if the mould merely consisted of boxes attached to a chain, very little of the coke made would be discharged from the boxes, even when they are completely inverted; in fact, it is extremely doubtful whether any of the blocks would fall out. Richards overcomes the difficulty by making the moulds as a part of the

chain itself. As each block of coke passes in its mould over the sprocket wheel at the discharge end of the conveyor, the sides of the mould, which are roughly at right angles to the direction of motion, alter their relative angle to one another, with the result that the carbonised block of coke is freed, and falls into the coke hopper. The methods discussed above are all typical of those in which the so-called " expansion " is allowed to take place, and mechanism introduced in order to set free the resultant product. It has already been shown that the expansion of coke may be reduced to zero by blending in suitable quantities a strongly swelling with a non-coking or contracting coal. Blending has been used for a great many years in the carbonising industries, particularly in coke oven practice. In the coke cakes made at H.M. Fuel Research Station's experimental setting, described on page 219, the mixture is adjusted so that there is a slight contraction when carbonisation is complete.

The laboratory assay method of Gray and King, developed at the Fuel Research Station,[1] provides a means for determining the most suitable proportions to give the desired result, a complete parallel having now been established between the assay and the large-scale working. This method of reducing the size of the bubble structure by blending is likely to bulk largely in any future carbonisation, since, if suitable coals are readily available, it provides a complete solution to one of the obstacles which baulked the earlier investigators, and renders it possible to utilise retorts of a much simpler form, and which are consequently cheaper in capital cost, upkeep and labour. A similar purpose may be effected by partial oxidation or by the pre-heat treatment process of Illingworth, which modifies the swelling tendency of a coking coal. All these methods have their utility, but the process of blending, provided always that both types of coal are available, presents considerable advantages from the national point of view, since it renders available a method for treating non-coking coals which often contain a considerable quantity of valuable oil-forming constituents. Blending also offers attractions to colliery owners, who sometimes find difficulty in disposing of non-coking slacks and smalls at other than very low prices.

D. DESIGN OF COMBUSTION CHAMBER AND HEATING ARRANGEMENTS

The apparatus for burning the fuel used in heating the setting forms a detail of the utmost importance in promoting the economy and the efficient working of externally heated retorts. Various types of fuels are available, ranging from solid, liquid, to gaseous. Of these the last-named, however, presents more advantages

---

[1] Fuel Research Board. Tech. Paper No. 1 : " The Assay of Coal for Carbonisation Purposes : a new Laboratory Method." Thos. Gray, D.Sc., Ph.D., and Jas. G. King, F.I.C., A.R.T.C. (H.M. Stationery Office.)

than either of the other two. With a gas fuel the utmost flexibility can be obtained, and since the development of high temperatures is unnecessary use can be made of a fuel of low calorific value, which can be cheaply obtained For this reason producer gas is undoubtedly likely to be the fuel utilised in the new industry if and when it becomes established. In the past use has often been made of the high calorific gas produced from the carbonisation process, but this is to be strongly deprecated. As we have already seen, this gas has intrinsically valuable qualities, and its sale under suitable conditions forms one of the least speculative assets of the process. Producer gas of satisfactory quality can be made from low-grade solid fuel, and indeed this is an economical method for utilising some of the breeze which will certainly be produced even in a well-designed plant.

Although a full consideration of the various methods whereby the potential heat of the fuel gas can be developed and transferred to the walls of the retort is beyond the scope of this volume, it is felt that a short discussion of the problem will be of advantage.

The producer itself may be one of many well-known types and need not be described. For information relating to the construction and method of working of producers the reader is referred to any standard book on the subject.

In some of the experimental plants for low temperature carbonisation which have been built or devised the design of the combustion chambers has been unduly complex and consequently expensive. In the simple case of a retort designed to heat the coal up to a temperature of 600° C. it is, generally speaking, quite unnecessary to provide a succession of heating chambers, each under separate control, such as is sometimes found desirable in high temperature practice. - In the Scottish shale industry the combustion chambers for oil distillation are of the simplest description, often consisting of one large chamber completely surrounding the retort, the gas being burned at the bottom of the chamber. Such a method has been adopted with great success in various types of low temperature heating furnaces at H.M. Fuel Research Station. Even with retorts of considerable height a very uniform temperature can be obtained from top to bottom. The cardinal principle which must be observed is to give the gas sufficient room to burn without impinging upon the metal of the retort. If the burning gas be made to travel upwards or downwards along the outer wall of the setting, the heat transfer to the retort itself will take place partly by radiation from the wall and partly by convection currents of hot gas. The combustion gas is supplied to the setting through ducts in the brickwork, suitable openings being provided above which combustion takes place. The air is supplied through separate ducts and openings, and if these are judiciously spaced with regard to the gas burners or openings the result is a quiet burning within the combustion chamber itself, giving the conditions which tend to a uniform temperature at all points in the chamber. The air openings must be capable of accurate

adjustment so that excess air may be reduced to a minimum and the temperature distribution kept under careful control.

In certain plants the gas has been burned in burners of the bunsen type, in which, as usual, the velocity of the entering gas is made to induce its own primary supply of air, arrangements being made to supply the secondary air as required. Such a method is absolutely unnecessary to effect the purpose desired. Indeed it is to be strongly deprecated, as it tends to localise excessively high temperatures and is also liable to give trouble through lighting back if the gas supply is not uniform.

It is difficult to give exact rules for the method which is here advocated, bu the following two examples of heating by this means are capable of modification to suit any particular case and may prove a useful guide to designers.

In Fig. 53 are shown the heating arrangements for the set of cast iron continuous vertical retorts designed for low temperature carbonisation of briquettes or certain types of coal at H.M. Fuel Research Station. The retorts are rectangular in shape with corners of large radius. At the lower end they are 2 ft. 9 in. by 1 ft. 3 in., tapering to 2 ft. 4 in. by 9 in. at the top. They are connected to a standard Glover-West extractor gear by the specially shaped connecting pieces shown. The four retorts are at present fed from a single hopper into which there is one opening at the centre ; the gas leaves each retort through an off-take pipe just below the hopper.

These retorts are set in a single heating chamber of which the brickwork is about 1 ft. 10½ in. in thickness, the inner 9 in. being of firebrick. As shown in the figure, the heating arrangement consists of four burners to each retort. The burners are placed in the corners of a rectangular space surrounding the base of the retort. The corners of this rectangle, where they are not formed by the actual corners of the main chamber, are built of brickwork, which extends horizontally into the chamber for a distance of 9 in., and vertically for a distance of 10 ft. 6 in. above the floor of the chamber. It is of great importance that the burners should be placed as close to each corner as possible. If placed further out the tendency is for the flames to leave the brickwork and impinge on the metal walls of the retort. The air supply for the setting is placed in a position midway between the various gas inlets. As a result of a series of experiments made on such heating arrangements, it was found that in this way the flames could be made to travel upwards in the corners without any considerable amount of eddying and without causing local overheating.

The space surrounding the hoppers is separate from that surrounding the retorts. A portion of the flue gases can, if necessary, be made to traverse this space in order to supply a controlled amount of heat to the hopper and so recover some of the sensible heat of the products of combustion. Ample provision has been made for placing

D

thermo-couples in various parts of the heating chamber. Those actually in use for experimental purposes are shown in the diagram, being numbered 1 to 8.

Table XLVII shows a series of determinations of temperature given by these thermo-couples with the retorts carbonising coal of comparatively low coking index

Section on A.A.

Plan showing positions for 4 Thermo Couples.
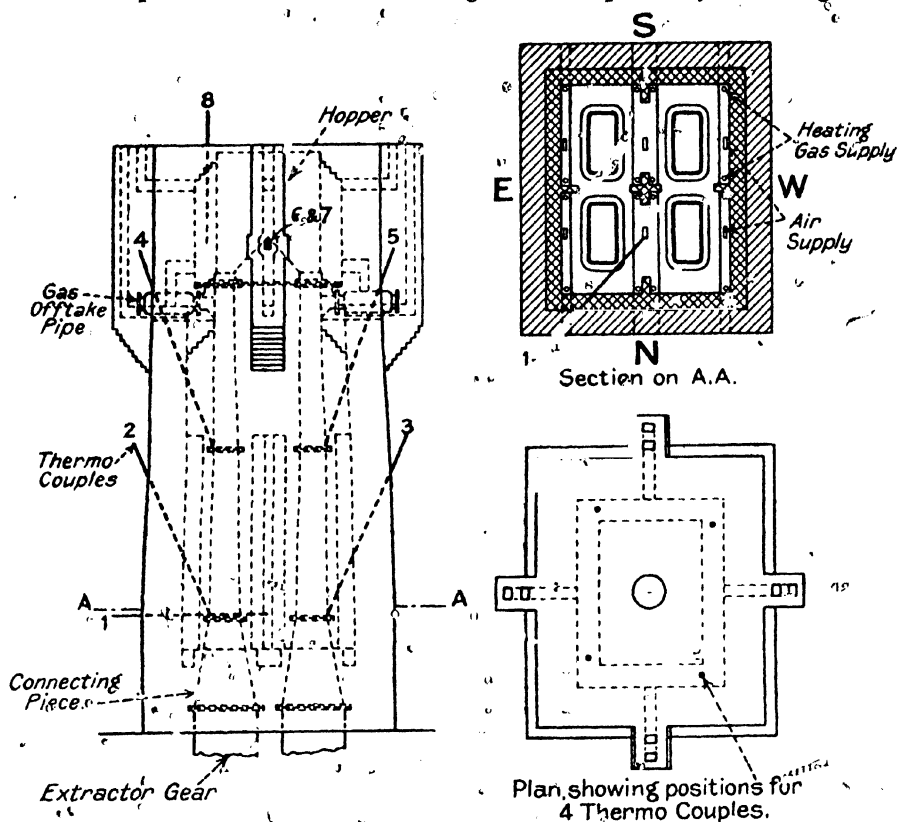
FIG. 63.—HEATING ARRANGEMENTS FOR LOW TEMPERATURE VERTICAL RETORT AT H.M. FUEL RESEARCH STATION.

and with the retorts filled with gas coke, the discharge arrangements being stopped in the second case. It will be seen that the first case gives therefore the temperatures obtained in continuous working, whilst the second corresponds to the case of an intermittently working retort. In the first case the maximum temperature of 606° C. is reached at the lower end, whilst at the flue at the top of the setting the temperature is only about 20° lower. In the case of intermittent working the general distri-

bution of temperature is not so uniform as in the first example, but on the north side the temperature of the metal at the bottom is only 21° higher than the temperature of the gas at the top of the setting, whilst on the south side the temperature of the metal is 38° above the temperature of the gas in the outlet flue. Such differences are not serious and could be considerably reduced by slight modification in the distribution of the gas supply.

The same principle is also illustrated in the heating of the horizontal retort setting described on page 220. In this case the gas and air are admitted through separate nostril blocks in the arch of the large heating chamber surrounding the retorts. The combustion of the gas is completed under the arch, which is maintained at a temperature considerably above 600° C. The arch radiates heat into the crown of the retorts and into the heating chamber, which is maintained thereby at a very uniform temperature throughout. Combustion is completed before the waste gases reach the retort so that the temperature of the metal is maintained with a minimum of action on the steel. The flue gases are led between the retorts to the floor of the setting and pass away by flues in the floor to short chimneys.

In this setting, with the exception of a slight fall in the temperature of the metal towards the front due to the heat drain caused by the doors of the retort, no difficulty has been experienced during long periods of working in maintaining an exceedingly uniform temperature within the carbonising chambers. The actual temperatures obtained in experimental working are given on page 25 of the Second Section of the Report of the Fuel Research Board for the years 1920–21.

TABLE XLVII. TEMPERATURES IN COMBUSTION CHAMBER OF VERTICAL RETORT AT H.M. FUEL RESEARCH STATION

No. of Couple.	Position.	Temperatures ° C.	
		Retorts working.	Retorts filled with Coke.
(1)	Horizontal at bottom N. side	597	572
(2)	Lower inclined E. side north position	599	566
(3)	,, ,, W. ,, south	606	583
(4)	Upper ,, E. ,, north	588	532
(5)	,, ,, W. ,, south	595	558
(6)	North flue at top of setting	584	545
(7)	Horizontal under centre of hopper (south)	464	480
(8)	Vertical in flue round hopper N.E. corner	327	446

E. SEPARATION OF TAR AND LIQUOR

An important point in connexion with a low temperature installation lies in the difficulty which is sometimes experienced in the separation of oil and liquor. In gasworks and coke ovens no particular troubles are associated with this, since the tar has a specific gravity sufficiently in excess of unity to ensure ready settling in the collecting tanks. With low temperature tar or oils, on the other hand, we have a material whose weight per unit volume is sometimes very little greater and sometimes even less than water. In many plants separation is only effected with difficulty.

In the tar made at H.M. Fuel Research Station, a large quantity of emulsified oil and liquor was found in the oil drawn from the collecting tanks and seals. Separation of the liquor, was, however, effected by passing the oil through a filter press, in order to extract the finely divided particles which tend to cause emulsification. After a further settling period of 48 hours the water content was reduced to below 3 per cent, and on further settling was reduced to below ½ per cent.

F. INTERNAL HEATING

We will now consider some of the troubles associated with the internal heating of retorts with hot gases. Many types have from time to time been suggested, and there are at least three which at the present time are attracting considerable attention. The advantages associated with this method of heating have already been referred to. With due care in the design of the apparatus, a uniform heat can be supplied practically simultaneously over the whole of any one zone of the apparatus. We are not therefore dependent upon a rise of temperature which passes slowly from the exterior to the interior, and which so seriously limits the bulk of coal which can be dealt with. Instead, it is possible to provide—as in the Maclaurin apparatus—a producer whose maximum diameter is 8 feet, with the accompanying advantages of large unit plant and the resulting saving in labour costs. The one defect of the process is the large quantity of gas which has to be dealt with; instead of 3,000–4,000 cubic feet per ton of rich gas, as much as 35,000 cubic feet of gas may be available, having a calorific value of say 230 B.Th.U. It has already been pointed out that the condensing and scrubbing plant has to be of sufficient size to cope with the increased volume, and also the well-known difficulty of scrubbing, and if necessary stripping a strongly diluted gas, makes for an increased size of plant. Again it is necessary to provide a market for the low-grade gas produced, which is much less likely to appeal to gas undertakings than is one of 900 B.Th.U. per cubic foot, this being of undoubted value for enrichment purposes. There is also no opening for the absorption of the low-grade gas as a fuel gas for heating the retorts themselves, as can be done, if necessary, in some of the other

processes. It would be necessary, therefore, to create a demand for this low-grade gas in the near neighbourhood of the plant. In spite, however, of the disabilities indicated, the method is a very attractive one, and in the authors' opinion retorts of this type are likely to take a prominent place in a low temperature carbonisation industry of the future.

An interesting development has already been mentioned which is taking place in Glasgow, where a five-unit battery of Maclaurin retorts is to be erected at Dalmarnock with a view to the production of solid smokeless fuel. In this case the problem of the proper utilisation of the gas has been solved by using it for boilers for the Electricity Department.

The supply by Gas Companies of a separate gas for power purposes has often been suggested, and is indeed foreshadowed, in the Fuel Research Board's report on Gas Standards.[1] Should this ever come about, an undoubted impetus would be afforded to the low temperature carbonisation by internal heating, which would supply an additional inducement to gas undertakings adopting, to some extent, a low temperature process, and so bringing the time nearer when such undertakings become universal suppliers of fuel as differentiated from their present outlook as suppliers of gas only as their main product, with the coke and tar as valuable by-products.

G. CARBONISATION IN SIMPLE VERTICAL RETORTS

The simplicity of a vertical Scottish shale retort has often led to the suggestion that carbonisation of bituminous coal might quite well be carried out in an apparatus of this type. The problems presented are mainly those already considered, and which are associated with difficulty in ensuring continuous working, owing to the fusing and swelling up of the material. A partial solution is presented by blending or pre-heat treatment, but unless the retorts are reduced to the narrowest dimensions, there will still be difficulty owing to the slowness with which heat can be made to travel through the coal. The series of experiments already referred to[2] in the carbonisation of coal in Glover West gas retorts proved that it was quite feasible to produce a good smokeless fuel with adequate recovery of oil and gas in such an apparatus. Troubles were, however, found in the tendency under certain conditions for coal to make its way through the centre core, and to arrive at the discharge end of the retort in a partially carbonised condition. Some improvement was effected by subjecting the charge to a light steaming process. The total volume of gas and vapour was increased thereby with the result that a better

[1] Fuel Research Board. Report to the Board of Trade on "Gas Standards." (Cmd. 108). (H.M. Stationery Office.)

[2] Fuel Research Board. Tech. Paper No. 7 : "Preliminary Experiments in the Low Temperature Carbonisation of Coal in Vertical Retorts." (H.M. Stationery Office.)

circulation of gas was secured within the retort, with a consequently improved distribution in the temperature conditions.

If we consider for a moment the case of the carbonisation of shale, we have a material which can be fed into the retorts in pieces of a fair size, which does not tend to stick together on heating, and in which the spent shale leaves the apparatus in the same form as it entered, so far as the shape and size of the lump are concerned. Besides having no tendency to agglomerate into unwieldy masses, the free convection which can be set up in such an open mass of material considerably increases the speed at which heat can be driven inwards to the centre of the change. The result is that the time of carbonisation is reduced, and greater uniformity of carbonisation is secured. With a non-coking coal of suitable size, similar conditions present themselves, and if by any process of treatment it is possible to convert a fusible bituminous coal into a material having the properties of shale enumerated above, many of the difficulties associated with retorting such a material will have been overcome. Several groups of workers are now engaged on experimental work in this direction. The Sutcliffe and Evans process already described is typical of the method. A mixture of swelling and non-swelling coal, or of a swelling coal and coke, is ground finely and briquetted cold, using a pressure of about 10 tons per square inch. With certain coals a robust briquette can be obtained which is sufficiently strong to withstand the subsequent treatment without the development of an undue amount of fines and breeze. The strength of the uncarbonised briquettes appears to depend to a considerable extent upon the degree of fineness to which the raw material is ground, and in an attempt to overcome the necessity for fine grinding and high briquetting pressures, the Fuel Research Board are experimenting with a method in which coal, which has merely been passed through a disintegrator, is heated to a temperature just short of the softening point. At this stage the material is briquetted at a pressure of about 2 tons per square inch, and then fed to the vertical cast iron retorts shown in Fig. 53, when carbonisation is effected at a temperature of about 600° C.

Another method for effecting the same purpose is to briquette a disintegrated coal of suitable composition with pitch, or other suitable binder, a proportion of the pitch being subsequently recovered in the retorting process. This method has been adopted in the process devised by Fisher and developed by the Midland Coal Products, Ltd., described on page 214. The question of pre-briquetting with subsequent carbonisation is one bristling with interesting possibilities, the work of Sutcliffe and Evans having shown that it is possible to produce certain modifications in the properties of cokes, which are likely to have considerable bearing on blast furnace practice.

The method developed by the Fuel Research Board of producing coke cakes by the carbonising of a suitable mixture of coking and non-coking coals in trays,

although originally developed entirely with a view to obtaining accurate data as
to the yields and products, presents certain possibilities, even in the commercial
direction. The products are satisfactory, and can be produced in the quality
required without waste or undue deterioration in the process. The retorts are
robust, easily worked, and are likely to have a long life if properly worked. The
disadvantage of the station setting lies in the labour cost, which is sufficiently
high to put the setting as at present designed out of court for commercial purposes
in this country. In countries, however, where labour is cheap and good prices
can be obtained for the smokeless fuel and oils produced, the system is worthy of
serious consideration. If a satisfactory method for the mechanical charging and
discharging of the trays can be developed, the method might have some application
in Great Britain. The difficulty associated with such a development of the station
setting is the comparatively complicated nature of the apparatus required. As
far as can be foreseen, the capital cost would be heavy, and the present investiga-
tions at H.M. Fuel Research Station are being pursued in directions which appear
at the moment to be more promising.

A development of the method of carbonisation in trays formed the subject of
experiments at the Fuel Research Station, with the retort described on page 221,
where the trays of crushed coal are superimposed one above the other in a series
of shelves which could be raised or lowered inside an iron retort. These shelves
were brought in turn in front of the charging and discharging opening, where they
were removed, emptied and discharged by suitable machinery. A large quantity
of good coke was made in this way, but the throughputs obtained were not such
as were likely to prove profitable in view of the somewhat elaborate mechanical
arrangements necessary.

• The method of superimposed trays has been adopted by Scott-Moncrieff in the
apparatus described on page 230. Here the charge is removed bodily from the
retorts when carbonisation is completed, and cooled in a cooling chamber, use
being made of the waste-heat of cooling. A very satisfactory smokeless fuel has
been obtained, but the plant has not as yet been worked for a sufficiently long
period to demonstrate whether a commercial success has been obtained.

CHAPTER XIX

THE ECONOMICS OF LOW TEMPERATURE CARBONISATION

INTRODUCTORY

A COMPLETE statement of the economics of low temperature carbonisation is very difficult at present. The matter has been before the public for a considerable time; but instead of the compilation of a wealth of reliable data from which balance sheets might be computed, we find a mass of conflicting figures, many of which are fundamentally unsound. So much prejudice has been aroused by the wild statements which have been issued from time to time, and which close analyses have revealed to be without foundation, that it may be said that these have had the result of hampering instead of assisting the prospect of establishing an industry capable of carbonising the tens of millions of tons of coal at present consumed in the raw state. Until recently no accurate data of yields from typical coals were generally available, with the result that erroneous figures, which were originally determined only by a few rough laboratory experiments, have been repeated again and again until the reader has become liable to accept these statements as accurate from sheer force of reiteration.

The establishment of a method of low temperature assay now enables the technician to determine accurately within a few hours the quantity and character of the products to be expected from the carbonisation of any given material. It is now possible to reject many figures quoted in the past which have exaggerated the yields of the more valuable products by so much as 150 per cent. Some of these figures carry with them their own refutation, as for example the series in which a fractionation of low temperature oil was made to yield in motor spirit, kerosene and Diesel oil the full quantity of the original oil, leaving entirely unaccounted for the losses on fractionating and also the pitch and other residues. Again, tar yields have frequently been measured as wet tar, thus exaggerating the true yields by the amounts of water present. In the estimates now submitted, which are framed on an assumption of the carbonisation of the whole of the domestic coal, it is not safe to take more than, say, 15 gallons per ton on the average.

ESTIMATES OF THE ASSETS OF THE PROCESS

It has been pointed out that the problem of low temperature carbonisation must be examined from two standpoints :, the technical and the economic. The data for the solution of the latter will only be obtained after the technical solution has been reached. In other words, until a fair-sized industrial plant has been operated continuously over a long period, manufacturing and disposing of all the products of carbonisation under steady market conditions, no one can say whether or not the business will be a profitable one. Although this stage does not appear

to have been reached, an attempt will be made to draw up tentative balance sheets.
In order to do this, assumptions will be made which can be modified if future
experience proves them to be incorrect.

We are faced at the outset by the uncertainty of market conditions, and by our
lack of knowledge as to the price at which low temperature coke will appeal to the
domestic fuel consumer. At the present time, the small quantities of fuel which
have from time to time been placed upon the market have commanded prices
which were often much in excess of those of coal, but it must be borne in mind
that such prices are unlikely to be maintained when a fair supply is obtainable,
and it is necessary to find customers other than those who are enthusiastic in their
support of any scheme which will tend to reduce the smoke evil in our large towns.
It is also essential that the consumer shall have ample assurance of being able to
obtain at any time supplies of the fuel, which must also be of fairly constant quality.
It has been considered in the past that it may be assumed that the price obtained
for the coke, as a high-class domestic fuel, will be sufficient to meet the entire cost
of the coal used. It is not desirable, however, to neglect the possibility that
smokeless fuel may only command a price equivalent to that of raw coal, weight
for weight. Since the yield of coke, per ton of coal, by straight low temperature
carbonisation amounts on the average to about 14 cwt., it will be seen, therefore,
that in the latter case the value of the liquid and gaseous products must be suffi-
cient to yield a profit after paying the entire costs of retorting and the cost of about
6 cwt. of the raw coal treated. Balance sheets, based on both assumptions, will be
given from which it will be seen that a process, which might be commercially sound
if the price of the coke is such as to pay the whole of the cost of the raw coal treated,
assumes a totally different aspect when the coke will only command a price
equivalent to raw coal, weight for weight.

Another difficulty, which lies in the way of drawing up even a very speculative
balance sheet, lies in the uncertainty as to the price which can be obtained for the
oils. This product will always have to face competition with imported oils, and the
price, therefore, in the absence of some measure of protection, is always dependent
upon factors outside the control of those interested in low temperature carbonisa-
tion. In this connexion, however, it has been found that several groups interested
in low temperature carbonisation have experienced no difficulty in disposing of
all the oil produced at a price of about 1s. a gallon, and this at a time when fuel
oil from foreign sources was available at a price in the neighbourhood of fourpence.
It is therefore evident that, for specific purposes, the oil has special properties
which are not possessed by natural fuel oils. Investigation of the oils carried out
by various workers has also shown that special products of various types are
available, and it is likely that markets may be created in the future for these
materials. In spite of this, however, such markets are not likely to be—nor indeed

is it conceivable that they should be,—in any way commensurate with the magnitude of a smokeless fuel industry, if once it were firmly established. The inevitable drop in oil prices would then have to be faced, to those corresponding to the largest and only reliable market—that of fuel oils of various types, the price of which is controlled in the main by foreign suppliers. Since the subject is dealt with in this volume from the national point of view, it is necessary to assume a price equivalent to that of crude fuel oil. In their recent computations, the Fuel Research Board have taken this as 5d. per gallon.

At this stage it may be mentioned that balance sheets of the type under discussion are frequently drawn up in terms of the fractionated products, the prices of which are of course considerably in excess of those of the crude oil from which they were obtained. For the present purpose, however, it is not desirable to do so, since this would involve the possible confusion between two distinct processes—those of retorting and refining of the crude oil—and complicating the issue by the necessity to debit the costs of the latter. It is also unjustifiable to go so far, since carbonisation must stand upon its own merits, without bringing to its aid the economics of another industry, possibly possessing an alternative source of supply for its raw material.

The remaining asset is the rich gas, of which about thirty therms are likely to be available. The Fuel Research Board have always emphasised the valuable nature of this product, which has frequently been credited as being merely the fuel necessary for retorting. The Board also point out the value of such gas to the gas undertakings as an enriching agent for gas of low thermal value, in order to help the undertakings to meet their obligations regarding the declared calorific value of the gas they supply. The enriching value of this gas is increased by the large amounts of methane and saturated hydrocarbons, which, in the large-scale tests at H.M. Fuel Research Station, varied between 41 and 52·4 per cent, according to the character of the particular coal or mixture of coals used. These hydrocarbons will not condense or separate out, either during storage or distribution, and on that account present advantages which are not possessed by some of the other gases and vapours used for enriching purposes at the present time. The gas is highly concentrated when considered from the standpoint of thermal value, and can accordingly be stored in relatively small holders and introduced at any moment into the district supply system. The Fuel Research Board have taken a tentative figure of 4d. per therm as the value of the gas for this purpose.

The only other by-product is the sulphate of ammonia, and it is frequently found that credits are taken for this which are hopelessly in excess of those likely to be realised. At the present time it would be wise to leave the value of this asset entirely out of the reckoning in the consideration of externally heated processes, owing to the uncertainty of the market and the low prices obtainable. In any case, as has already been pointed out, the quantity likely to be produced is small.

ESTIMATES OF THE COSTS OF THE PROCESS

Having now arrived at some basis for the items to be entered to the credit side of the balance sheet, the debit side remains to be considered.

The cost of the material to be carbonised forms one of the principal items. Here it is necessary again to lay stress upon a point of fundamental importance in the consideration of the subject, i.e. whether the project is for the moment under consideration from the national point of view, or from the standpoint of some particular scheme of more local significance. An examination of the subject in the broadest sense reveals the fact that the price of the raw material is likely to be the same, generally speaking, as that obtainable when sold as domestic or industrial fuel. In other words, if the greatest ultimate benefit is to be obtained from low temperature carbonisation, it will be necessary to carbonise practically the whole of the coal at present used for domestic purposes. When, however, the subject is considered in connexion with the utilisation of slack and small coals, which at present are of the nature of waste products, it is possible to debit the fuel to the plant at a low price. Especially would this be the case where—as might happen in many instances—the firm working the plant were the colliery company interested in the disposal of a comparatively low-priced product. If the carbonising plant were separately owned, however, the outlook might not be so favourable, since a multiplication of such plants would inevitably result in an increase in price of the raw material, with the consequent transfer of some of the profits to the coal owner. The law of supply and demand would of course operate, and would tend to the establishment of prices at which the infant carbonising industry could barely exist. Unless, however, a design of plant and a process can be evolved which will be operated sufficiently cheaply to enable the full economic cost of the raw material to be priced as a main product of a colliery company, and not as one of its waste products, it will be impossible to establish the new industry on a scale commensurate with the needs of the nation as a whole.

Sir George Beilby [1] has estimated the capital expenditure required for the installation of plant capable of carbonising 35 million tons of coal per annum as about 30 to 40 millions sterling. Taking the average of these figures, the capital cost for each ton carbonised, per annum, would be of the order of £1. Such a figure would be applicable to a fair-sized plant having a throughput of, say, 65,000 tons per annum, and each of the following estimates is based on a plant of about this capacity.

The entire pre-war cost of carbonisation in the Scottish shale industry was about 1s. 6d. per ton, including labour, maintenance and fuel for heating. The

[1] Beilby, Sir George, *Fuel Problems of the Future*. Inst. of Civil Engineers: James Forrest Lecture.

modern equivalent of this figure would be about 3s. per ton, but in the following estimates 4s. has been taken for this item.

TENTATIVE BALANCE SHEETS

On the basis of the estimates put forward in the two preceding paragraphs, balance sheets are submitted to indicate the financial possibilities of the undertaking. Balance sheets No. 1 and No. 3 are drawn up on the assumption that the coke made pays for the whole of the coal used, and that this coal can be delivered in bulk to the plant at 30s. per ton and 20s. per ton respectively. Balance sheets No. 2 and No. 4 correspond to the cases where the coke at the plant will only command the price of raw coal, weight for weight.

BALANCE SHEET No. 1

Income	£	s.	d.	Expenditure	£	s.	d.
Coke, 14 cwt. (i.e. 42s. 10d. per ton)	1	10	0	Coal, 1 ton	1	10	0
Oil, 15 gall. @ 5d.		6	3	Cost of carbonising (labour, maintenance and fuel for heating)		4	0
Gas, 30 therms @ 4d.		10	0				
				Interest and depreciation, say 20 per cent		4	0
	2	6	3		1	18	0

Balance : 8s. 3d. per ton of coal carbonised, or 41 per cent on cost of plant.

BALANCE SHEET No. 2

Income	£	s.	d.	Expenditure	£	s.	d.
Coke, 14 cwt. @ 1s. 6d. (i.e. 30s. per ton)	1	1	0	Coal, 1 ton	1	10	0
Oil, 15 gall. @ 5d.		6	3	Cost of carbonising (labour, maintenance and fuel for heating)		4	0
Gas, 30 therms @ 4d.		10	0				
				Interest and depreciation, say 20 per cent		4	0
	1	17	3		1	18	0

Balance : 9d. adverse.

BALANCE SHEET No. 3

Income		£	s.	d.	Expenditure		£	s.	d.
Coke, 14 cwt. (i.e. 28s. 7d. per ton)	.	1	0	0	Coal, 1 ton . . .		1	0	0
Oil, 15 gall. @ 5d.	.		6	3	Cost of carbonising (labour, maintenance and fuel for heating)	.		4	0
Gas, 30 therms @ 4d.	.		10	0	Interest and depreciation, say 20 per cent	. .		4	0
		1	16	3			1	8	0

Balance : 8s. 3d. per ton of coal carbonised, or 41 per cent on cost of plant, which of course is identical with that of Balance Sheet No. 1.

BALANCE SHEET No. 4

Income		£	s.	d.	Expenditure		£	s.	d.
Coke, 14 cwt. @ 1s. (i.e. 20s. per ton)	. .		14	0	Coal, 1 ton . . .		1	0	0
Oil, 15 gall. @ 5d.	.		6	3	Cost of carbonising (labour, maintenance and fuel for heating)	. .		4	0
Gas, 30 therms @ 4d.	.		10	0	Interest and depreciation, say 20 per cent	.		4	0
		1	10	3			1	8	0

Balance : 2s. 3d. per ton, or 11 per cent on cost of plant.

ESTIMATED BALANCE SHEET FOR COKE OVEN PLANT

Although not strictly comparable, it will be of interest to compare the above figures with the estimates which have been furnished to the Authors for a coke oven plant now being erected in the North of England, and capable of carbonising 65,000 tons of coal per annum.

In this case, the raw material is supplied in the form of unwashed slack at a price of 13s. 6d. per ton. The capital cost of the installation would be about £100,000, including plant, buildings, foundations and sidings. This is equivalent to 30s. for each ton carbonised per annum.

BALANCE SHEET FOR COKE OVEN PLANT

Income	£	s.	d.	Expenditure	£	s.	d.
Coke, 13·2 cwt. @ 25s. per ton		16	5	Unwashed slack, 1 ton @ 13s. 6d.		13	6
Tar, 8 gall. @ 5d.		3	4	Cost of carbonising, oven to benzol stills, including materials, stores, acid, labour, etc., 5s. per ton		5	0
Sulphate of ammonia, 33 lb. @ 1½d.		4	1				
Crude benzol, 3 gall. @ 9d.		2	3				
Surplus gas, 5,000 cu. ft. @ 6d. per 1,000 cu. ft.		2	6	5 per cent on cost of plant		1	6
				10 per cent amortisation		3	0
	1	8	7		1	3	0

Balance : 5s. 7d. per ton, or 18·6 per cent on cost of plant, etc.

ESTIMATED BALANCE SHEET FOR MACLAURIN PRODUCER

A plant of the Maclaurin type is exceedingly interesting, and presents great possibilities owing to its low capital cost. Its success, however, depends very largely upon the securing of a profitable outlet for the large quantities of gas of about 230 B.Th.U. per cubic foot. It is obvious that it is only useful as a fuel gas for industrial purposes. Mr. Maclaurin, in his own estimates, has taken the value of the gas as varying from 1·8 to 2·3d. per therm, the former figure representing its value when used for steam-raising in comparison with coal at 20s. per ton. He also estimates the capital cost of a battery of 10 units, capable of a throughput of 200 tons of coal per day, as £12,000 for producers alone at present-day prices. With all accessories, such as elevators, conveyors, motors, turbines, fans, pumps, storage bunkers and tanks, buildings and piping, the plant would, in his opinion, cost about £30,000. The capital cost therefore works out at about 10s. for each ton of coal treated per annum.

The following balance sheet has therefore been worked out from the figures supplied by the Maclaurin Company. The coke is assumed to command the same price per ton as that given in Balance Sheet No. 3, the plant being situated in a district where suitable coal is delivered in bulk at 20s. per ton. On this basis, a price of 42s. 10d. per ton would be obtained for the smokeless fuel.

BALANCE SHEET FOR MACLAURIN PRODUCER MAKING SMOKELESS FUEL

Income	£	s.	d.	*Expenditure*	£	s.	d.
Coke, 11 cwt. @ 12s. 10d. per ton . . .				Coal, 1 ton . . .	1	10	0
	1	3	7	Interest, repairs and depreciation, 25 per cent		2	6
Oil, 15 gall. @ 5d. . .		6	3				
Sulphate of ammonia, 17 lb. @ ¾d.		1	1	General charges, stores, rates, etc.		2	0
Gas, 68 therms @ 1·8d.		10	2	Steam			6
				Labour and supervision .		2	0
	2	1	1		1	17	0

Balance : 4s. 1d. per ton, or 41 per cent on cost of plant.

CONCLUSIONS

If the various assumptions regarding capital cost and the prices obtainable for by-products are substantially correct, and if the technical difficulties of the process can be successfully overcome, it will be seen that the commercial success of a plant will be mainly dependent on the ratio which the price commanded by the coke bears to the cost of the raw coal. If it can be sold at a price only equivalent, weight for weight, with that of the raw coal, the prospect of establishing an industry commensurate with our national requirements is by no means promising. On the other hand, if the raw material can be obtained at a price such that the coke made will approach in value the whole cost of the coal used, there would appear to be a sufficient margin for commercial success. This, however, can only be definitely settled when an industrial plant has been running continuously for a considerable period, the products being marketed at a fair price and yielding a reasonable profit to the shareholders.

The considerations already referred to form the only safe basis on which the business man can estimate the probabilities of ultimate success held out by any suggested scheme. In consideration of the problem from the national standpoint, however, it is important not to lose sight of the considerable benefits and indirect savings which would follow the use of smokeless fuel on an adequate scale. In a previous chapter, the direct loss accruing both to the individual and the nation was discussed. The taking of such savings into consideration can only be effected by Parliamentary action in the direction of some kind of subsidy or preference to smokeless fuel. How such a subsidy could be granted is a question of much difficulty. Enthusiasts have suggested such drastic enactments as the total prohibition of raw coal burning. When the innate conservatism of the British public is taken into

consideration, however, it will be admitted that such action could only follow the education of the public in the manifest advantages offered. Excellent work in this direction is going forward, and the value of the labours of the many organisations which are interesting themselves in the matter cannot be over-estimated should their labours result in the willingness of the public to show their appreciation of the advantages of smokeless fuel to the extent of paying a price for the coke which would render the process economically sound, Parliamentary action would be unnecessary. In either case, the alleviation of the smoke nuisance, and the provision of home supplies of oil, whether by the method discussed in this volume, or by any other sound method, can only be achieved by the enlightenment of the consumer in the direction of his best interests.

INDEX TO NAMES OF AUTHORITIES